Anomalous Argon-Hydrogen-Strontium Discharge

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We report the observation of intense extreme ultraviolet (EUV) emission from incandescently heated atomic hydrogen and atomized strontium that increased with argon. Typically the emission of extreme ultraviolet light from hydrogen gas is achieved via a discharge at high voltage, a high power inductively coupled plasma, or a plasma created and heated to extreme temperatures by RF coupling (e.g. $> 10^6 \ K$) with confinement provided by a toroidal magnetic field. The observed plasma formed at low temperatures (e.g. $\approx 10^3 \ K$) from atomic hydrogen generated at a tungsten filament that heated a titanium dissociator and atomic strontium which was vaporized from the metal by heating. The emission intensity of the plasma generated by atomic strontium increased significantly with the introduction of argon gas only when Ar^+ emission was observed. No emission was observed with hydrogen when sodium, magnesium, or barium replaced strontium or with hydrogen, hydrogen-argon mixtures, or strontium alone. The power balance of a gas cell having vaporized strontium and atomized hydrogen from pure hydrogen or argon-hydrogen mixture (77/23%) was measured by integrating the total light output corrected for spectrometer system response and energy over the visible range. Hydrogen control cell experiments were identical except that sodium, magnesium, or barium replaced strontium. In the case of hydrogen-sodium, hydrogen-magnesium, and hydrogen-barium mixtures, 4000, 7000, and 6500 times the power of the hydrogen-strontium mixture was required, respectively, in order to achieve that same optically measured light output power. With the addition of argon to the hydrogen-strontium plasma, the power required to achieve that same optically measured light output power was reduced by a factor of about two. The power required to maintain a plasma of equivalent optical brightness with strontium atoms present was 8600 and 6300 times less than that required for argon-hydrogen and argon control, respectively. A plasma formed at a cell voltage of about 250 V for hydrogen alone and sodium-hydrogen mixtures, 140-150 V for hydrogen-magnesium and hydrogen-barium mixtures, 224 V for an argon-hydrogen mixture, and 190 V for argon alone; whereas, a plasma formed for hydrogen-strontium mixtures and argon-hydrogen-strontium mixtures at extremely low voltages of about 2 V and 6.6 V, respectively.

I. INTRODUCTION

A historical motivation to cause EUV emission from a hydrogen gas was that the spectrum of hydrogen was first recorded from the only known source, the Sun [1]. Developed sources that provide a suitable intensity are high voltage discharge, synchrotron, and inductively coupled plasma generators [2]. An important variant of the later type of source is a tokomak [3] wherein a plasma is created and heated to extreme temperatures by RF coupling (e.g. $>10^6 K$) with confinement provided by a toroidal magnetic field. In contrast, it has been reported that intense EUV emission was observed at low temperatures (e.g. $\approx 10^3 K$) from atomic hydrogen and certain atomized elements or certain gaseous ions which singly or multiply ionize at integer multiples of the potential energy of atomic hydrogen, $27.2\ eV$ [4-18]. The ionization energy of Ar^+ to Ar^{2+} is $27.6\ eV$ [19], and an electric field may adjust the energy of ionizing Ar^+ to Ar^{2+} to match the energy of $27.2\ eV$. Thus, a discharge of argon is anticipated to cause an anomalous discharge when hydrogen is present.

Kuraica and Konjevic [20] observed intense wing developments of hydrogen Balmer lines with argon present in the negative glow of a glow discharge of an argon-hydrogen mixture irrespective of cathode material (carbon, copper, and silver). An anomalous discharge was not observed in neon-hydrogen and pure hydrogen mixtures. The authors offer a tentative explanation for hydrogen line shapes in the presence of argon which is based on a quasiresonace charge transfer between metastable argon ions and hydrogen molecules and the formation of a hydrogen molecular ion. According to the authors,

"... it is essential that the H_2^+ or H_3^+ ion must gain energy in the electric field before dissociation. Otherwise, the large energy of excited hydrogen atoms (on the average 50 eV per atom) cannot be explained".

The source of $50\ eV$ anomalous thermal broadening of the Balmer lines observed by Kuraica and Konjevic [20] may be dependent on the presence of an atom or ion which ionizes at about an integer multiple of

27.2 eV since Ar^+ ionizes at 27.6 eV.

Strontium ionizes at integer multiples of the potential energy of atomic hydrogen; thus, a hydrogen-strontium mixture was tested for anomalous EUV emission and plasma formation relative to mixtures of hydrogen and chemically similar controls that do not have electron ionization energies which are a multiple of 27.2 eV.

We report that a hydrogen plasma is formed at low temperatures (e.g. $\approx 10^3 K$) by reaction of atomic hydrogen with strontium atoms, but not with magnesium, barium, or sodium atoms. In the case of EUV measurements, atomic hydrogen was generated by dissociation at a tungsten filament and at a transition metal dissociator that was incandescently heated by the filament. Strontium atoms were vaporized by heating to form a low vapor pressure (e.g. 1 $torr$). The kinetic energy of the thermal electrons at the experimental temperature of $\approx 10^3 K$ were about 0.1 eV, and the average collisional energies of electrons accelerated by the field of the filament were less than 1 eV. (No blackbody emission was recorded for wavelengths shorter than 400 nm.). Strontium atoms caused hydrogen EUV emission; whereas, the chemically similar atoms, sodium, magnesium, and barium, caused no emission. The emission intensity of the plasma generated by atomic strontium increased significantly with the introduction of argon gas only when Ar^+ emission was observed. No emission was observed with argon or argon-hydrogen mixtures without strontium.

Furthermore, an anomalous hydrogen-strontium discharge was observed by visible emission. A cylindrical nickel mesh hydrogen dissociator of a gas cell also served as an electrode to produce an essentially uniform radial electric field between the dissociator and the wall of the cylindrical stainless steel gas cell. Power was applied to the electrode to achieve a bright plasma which was recorded over the wavelength range $350 \leq \lambda \leq 750\, nm$. The power balance of a gas cell having atomized hydrogen and strontium was measured by integrating the total light output corrected for spectrometer system response and energy over the visible range. Control experiments were identical except that sodium, magnesium, or barium replaced strontium. In the case of hydrogen-sodium, hydrogen-magnesium, and hydrogen-barium mixtures, 4000, 7000, and 6500 times the power of the hydrogen-strontium mixture was

required, respectively, in order to achieve that same optically measured light output power. With the addition of argon to the hydrogen-strontium plasma, the power required to achieve that same optically measured light output power was reduced by a factor of about two. In the case of an argon-hydrogen mixture and argon alone, the power requirement was 8600 and 6300 times the power input of the argon-hydrogen-strontium mixture, respectively.

II. EXPERIMENTAL

A. EUV spectroscopy

Due to the extremely short wavelength of this radiation, "transparent" optics do not exist for EUV spectroscopy. Therefore, a windowless arrangement was used wherein the source was connected to the same vacuum vessel as the grating and detectors of the EUV spectrometer. Windowless EUV spectroscopy was performed with an extreme ultraviolet spectrometer that was mated with the cell. Differential pumping permitted a high pressure in the cell as compared to that in the spectrometer. This was achieved by pumping on the cell outlet and pumping on the grating side of the collimator that served as a pin-hole inlet to the optics. The cell was operated under gas flow conditions while maintaining a constant gas pressure in the cell. The gas pressure inside the cell was maintained at about 300 mtorr with a hydrogen flow rate of 5.5 sccm controlled by a 20 sccm range mass flow controller (MKS 1179A21CS1BB) with a readout (MKS type 246). The argon-hydrogen gas mixture which produced the maximum EUV emission was determined by adjusting the flow rate of hydrogen and argon with two mass flow controllers.

The experimental set up shown in Figure 1 comprised a quartz cell which was 500 mm in length and 50 mm in diameter. Three ports for gas inlet, outlet, and photon detection were on a Pyrex cap of the cell that sealed to the quartz cell with a Viton O ring and a C-clamp. A tungsten filament (0.508 mm in diameter and 800 cm in length, total resistance ~2.5 ohm) heater and hydrogen dissociator and in the case of the hydrogen gas experiments, a titanium cylindrical screen (300 mm long

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and 40 mm in diameter) that performed as a second hydrogen dissociator were inside the quartz cell. A new dissociator was used for each hydrogen gas experiment. The filament was coiled on a grooved ceramic tube support to maintain its shape when heated. The return lead passed through the inside of the ceramic tube. The titanium screen was electrically floated. The power was applied to the filament by a Sorensen 80-13 power supply which was controlled by a constant power controller. The temperature of the tungsten filament was estimated to be in the range of 1100 to 1500 °C. The external cell wall temperature was about 700 °C. The entire quartz cell was enclosed inside an insulation package comprised of Zircar AL-30 insulation. Several K type thermocouples were placed in the insulation to measure key temperatures of the cell and insulation. The thermocouples were read with a multichannel computer data acquisition system.

In the present study, the light emission phenomena was studied for 1.) hydrogen, argon, neon, and helium alone; 2.) sodium, magnesium, barium, and strontium metals alone; 3.) sodium, magnesium, barium, and strontium with hydrogen; and 4.) sodium, magnesium, barium, and strontium with an argon-hydrogen mixture (97/3%). The pure elements of magnesium, barium, and strontium were placed in the bottom of the cell and vaporized by the filament heater. The power applied to the filament was 300 W in the case of strontium and up to 600 watts in the case of magnesium, barium, and sodium metals. The voltage across the filament was about 55 V and the current was about 5.5 ampere at 300 watts. For the controls, magnesium, barium, and sodium metals, the cell was increased in temperature to the maximum permissible with the power supply.

The light emission was introduced to an EUV spectrometer for spectral measurement. The spectrometer was a McPherson 0.2 meter monochromator (Model 302, Seya-Namioka type) equipped with a 1200 lines/mm holographic grating with a platinum coating. The wavelength region covered by the monochromator was $30-560\,nm$. A channel electron multiplier (CEM) was used to detect the EUV light. The wavelength resolution was about $1\,nm$ (FWHM) with an entrance and exit slit width of $300\,\mu m$. The vacuum inside the monochromator was maintained below 5×10^{-4} torr by a turbo pump. The EUV spectrum

$(40-160\,nm)$ of the cell emission with strontium present was recorded at about the point of the maximum Lyman α emission.

The UV/VIS spectrum $(40-560\,nm)$ of the cell emission with hydrogen alone was recorded with a photomultiplier tube (PMT) and a sodium salicylate scintillator. The PMT (Model R1527P, Hamamatsu) used has a spectral response in the range of $185-680\,nm$ with a peak efficiency at about $400\,nm$. The scan interval was $0.4\,nm$. The inlet and outlet slit were $500\,\mu m$ with a corresponding wavelength resolution of $2\,nm$.

B. Power cell apparatus and procedure

Plasma studies with 1.) hydrogen, argon, or argon-hydrogen mixture alone, 2.) hydrogen with strontium, sodium, magnesium, or barium, and 3.) argon-hydrogen mixture (77/23%) with strontium were carried out in the cylindrical stainless steel gas cell shown in Figure 2. The experimental setup for generating a glow discharge hydrogen plasma and for optically measuring the power balance is shown in Figure 3. The cell was heated in a 10 kW refractory brick kiln (L & L Kiln Model JD230) as shown in Figure 3. The cell was evacuated and pressurized with hydrogen, argon, or argon and hydrogen through a single 0.95 cm feed through. The discharge was started and maintained by an alternating current electric field in the 1.75 cm annular gap between an axial electrode and the cell wall. The cylindrical cell was 9.21 cm in diameter and 14.5 cm in height. The axial electrode was a 5.08 cm OD by 7.2 cm long stainless steel tube wound with several layers of nickel screen. The overall diameter of the axial electrode was 5.72 cm. A 1.6 mm thick UV-grade sapphire window with 1.5 cm view diameter provided a visible light path from inside the cell. The viewing direction was normal to the cell axis. A 1.27 cm diameter stainless steel tube passed through the furnace wall and connected to a view port welded to the cell wall at mid-height to provide an optical light path from the sapphire window to the furnace exterior. An 8 mm quartz rod channeled the light from the view port through the stainless tube to a collimating lens which was focused on a 100 μm optical fiber located outside the furnace. Spectral data was recorded with a visible spectrometer (Ocean Optics S2000) and stored by

a personal computer.

The field voltage was controlled by a variable voltage transformer operating from 115 VAC, 60 Hz. A step-up transformer was used when necessary. True rms voltage at the axial electrode was monitored by a digital multimeter (Fluke 8010 A or Tenma 726202). A second multimeter (Extech 380763) in series with the discharge gap was used to indicate the current. The cell temperature was measured by a thermocouple probe located in the cell interior approximately 2 cm from the discharge gap. The pressure in the hydrogen and argon supply tube outside the furnace was monitored by 10 torr and 1000 torr MKS Baratron absolute pressure gauges. In the absence of gas flow, the gas supply tube pressure was essentially the cell pressure. The pressure of each gas in an argon-hydrogen mixture was determined by adding one pure gas to a given pressure and increasing the pressure with a second gas to a final pressure. The partial pressure of the second gas was given by the incremental increase in total gas pressure.

Strontium (Aldrich Chemical Company 99.9 %), sodium (Aldrich Chemical Company 99.95 %), magnesium (Alfa Aesar 99.98 %), or barium (Aldrich Chemical Company 99.99 %) metal was loaded into the cell under a dry argon atmosphere. The cell was evacuated with a turbo vacuum pump to a pressure of 4 mtorr during most of the heating process. During the heat-up the cell was periodically pressurized with hydrogen (99.999% purity) to approximately 100 torr and subsequently evacuated to purge gaseous contaminants from the system. When the cell temperature stabilized hydrogen was added until the steady pressure was approximately 1 torr. The field voltage was increased until breakdown occurred. This was confirmed by the spectrometer response to visible light emitted from the cell. The hydrogen or hydrogen-argon pressure was adjusted, as much as possible, to maximize the light emission from the cell. The voltage was maintained at the minimum level which resulted in a stable discharge during data acquisition.

The spectrometer system comprised a 100 µm optical fiber and visible spectrometer (Ocean Optics S2000). To correct for the nonuniform response of the spectrometer system as a function of wavelength and the dependence of energy on wavelength, the system was calibrated against a reference light source (Ocean Optics LS-1-CAL). A spectral calibration

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factor was applied to the count rate data at each wavelength to yield the irradiation of the detector in units of energy/time/area/wavelength. The total visible radiant flux incident on the detector was calculated by integrating the spectral irradiation between 400 and 700 nm.

III. RESULTS

A. EUV spectroscopy

The cell without any test material present was run to establish the baseline of the spectrometer. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, and 300 mtorr hydrogen with a flow rate of 5.5 sccm is shown in Figure 4. The corresponding UV/VIS spectrum ($40-560\ nm$) is shown in Figure 5. The spectrum was recorded with a photomultiplier tube (PMT) and a sodium salicylate scintillator. No emission was observed except for the blackbody filament radiation at the longer wavelengths. No emission was also observed from the pure elements alone or when argon, neon, or helium replaced hydrogen.

The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized sodium, magnesium, or barium metal, and 300 mtorr hydrogen with a flow rate of 5.5 sccm are shown in Figures 6, 7, and 8, respectively. Sodium, magnesium, or barium metal was vaporized by filament heating. No emission was observed in any case. The maximum filament power was greater than 500 watts. At 500 watts, the temperature of the magnesium was 1000 °C which would correspond to a vapor pressure of about 100 mtorr. A metal coating formed in the cap of the cell over the course of the experiment in each case.

The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, a vaporized strontium metal, and 300 mtorr hydrogen with a flow rate of 5.5 sccm is shown in Figure 9. Strong emission was observed from vaporized strontium and hydrogen. The

EUV spectrum (40−160 nm) of the cell emission recorded at about the point of the maximum Lyman α emission is shown in Figure 10. No emission was observed in the absence of hydrogen flow. A metal coating formed in the cap of the cell over the course of the experiment.

Hydrogen was replaced by a 97% argon and 3% hydrogen mixture. The intensity of the Lyman α emission as a function of time from each gas cell was recorded. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a CEM is shown in Figure 11. No emission was observed with argon-hydrogen mixtures alone. The EUV spectrum (40−160 nm) of the sodium, magnesium, and barium gas cell emission that was recorded with a PMT and a sodium salicylate scintillator two hours after the cell reached 700 °C are shown in Figures 12, 13, and 14, respectively. Each cell comprised a tungsten filament, vaporized metal, and 300 mtorr argon-hydrogen mixture (97/3%). No emission was observed in any case.

The EUV spectrum (40−160 nm) of the cell emission recorded at about the point of the maximum Lyman α emission from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, vaporized strontium metal, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a PMT and a sodium salicylate scintillator is shown in Figure 15. Strong EUV emission was observed that was more intense than with hydrogen and strontium. The Lyman series corresponded to atomic hydrogen emission and strong Ar^+ ion emission was observed at 92.0 nm and 93.2 nm.

The anomalous plasma required the presence of hydrogen. It was found that increasing the hydrogen pressure initially increased the atomic hydrogen emission lines, but with increasing hydrogen partial pressure at a constant total pressure, the Ar^+ emission in the EUV at 92.0 nm and 93.2 nm decreased which resulted in a decrease of the plasma intensity including the hydrogen emission. The zero order emission in the EUV was observed with titration of increasing partial pressure of hydrogen added to argon gas. The optimum argon-hydrogen gas mixture which produced the greatest emission was determined to be 95% argon and 5% hydrogen which is similar to the 97% argon and 3% hydrogen

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mixture of Kuraica and Konjevic [20]. The optimum ratio was consistent with an anomalous discharge mechanism which required maximum concentrations of both atomic hydrogen and Ar^+.

B. Optically measured power balance

Count rate and spectrometer system irradiation of the background spectrum of hydrogen and strontium vapor over the wavelength range $350 \leq \lambda \leq 750\, nm$ in the absence of power applied to the electrode and in the absence of a discharge is shown in Figure 16. This data was collected during cell evacuation following the test with strontium and hydrogen at a cell temperature of 664 °C. The maximum visible irradiation of $0.004\, \mu W/cm^2 nm$ occurred at the red end of the visible spectrum. The results are summarized in Table I where T is the temperature, P_{hyd} and P_{Ar} are the hydrogen and argon partial pressures, and P_v is the equilibrium metal vapor pressure calculated from standard curves of the vapor pressure as a function of temperature [21].

Power was applied to the electrode to achieve a bright plasma in the strontium-hydrogen mixture and the controls of hydrogen alone, and sodium-hydrogen, magnesium-hydrogen, and barium-hydrogen mixtures for cell temperatures in the range 335-666 °C. In each case, the spectral radiant flux at the spectrometer system was recorded. If possible, the power driving the controls was adjusted such that the peak spectrometer system spectral irradiation was about $0.1\, \mu W/cm^2 nm$ in each case. The integrated visible irradiation levels were of the order of $1\, \mu W/cm^2$. One exception was the case of hydrogen-barium. In this case, the maximum spectral irradiation levels and integrated visible irradiation levels were only of the order of $0.01\, \mu W/cm^2 nm$ and $0.03\, \mu W/cm^2$, respectively.

The power required to maintain a plasma of equivalent optical brightness with strontium atoms present was 4000, 7000, and 6500 times less than that required for the sodium, magnesium, and barium control, respectively. A driving power of 33.7 W and 58 W was necessary to achieve a total visible radiant flux of about $1\, \mu W/cm^2$ from a sodium-hydrogen mixture and a magnesium-hydrogen mixture, respectively. For a hydrogen-barium mixture, a power input of about 55 W was required to achieve a total visible irradiation of about $0.03\, \mu W/cm^2$.

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Whereas, in the case of a strontium-hydrogen mixture, a power input of 8.5 mW resulted in a plasma with a total visible radiant flux of about the same optical brightness as sodium and magnesium. A plasma formed at a cell voltage of about 250 V for hydrogen alone and sodium-hydrogen mixtures, and 140-150 V for hydrogen-magnesium and hydrogen-barium mixtures; whereas, a plasma formed for hydrogen-strontium mixtures at the extremely low voltage of about 2 V. The results are summarized in Table I.

Power was applied to the electrode to achieve a bright plasma in the strontium-argon-hydrogen mixture and the controls of argon alone and argon-hydrogen alone for cell temperatures in the range 514-520 °C. In each case, the spectral radiant flux at the spectrometer system was recorded. If possible, the power driving the controls was adjusted such that the peak spectrometer system spectral irradiation was about $0.1 \, \mu W/cm^2 nm$ in each case. The integrated visible irradiation levels were of the order of $1 \, \mu W/cm^2$.

The power required to maintain a plasma of equivalent optical brightness with strontium atoms present was 8600 and 6300 times less than that required for argon-hydrogen and argon control, respectively. A driving power of 33.5 W and 24.7 W was necessary to achieve a total visible radiant flux of about $1 \, \mu W/cm^2$ from an argon-hydrogen mixture and argon, respectively. Whereas, in the case of a strontium-argon-hydrogen mixture, a power input of 4 mW resulted in a plasma with a total visible radiant flux of about the same optical brightness as the argon-hydrogen mixture and argon. A plasma formed at a cell voltage of 224 V for an argon-hydrogen mixture alone, and 190 V for argon alone; whereas, a plasma formed for argon-hydrogen-strontium mixtures at an extremely low voltage of about 6.6 V. The results are summarized in Table I.

The count rate and the spectrometer system irradiation for a mixture of hydrogen and strontium vapor at 664 °C is shown in Figure 17. Optimal light emission was observed after several hours of cell evacuation. The hydrogen partial pressure was unknown under these conditions. The calculated equilibrium vapor pressure of strontium at 664 °C is approximately 270 mtorr. The measured breakdown voltage was approximately 2 V. The maintenance voltage for a stable discharge

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was 2.2 V and input power was 8.5 mW. Spectral characteristics are noted in Table II. The hydrogen Balmer α and β peaks were obscured by strong strontium emission near 654.7 and 487.2 nm, respectively.

The spectrometer system irradiation for a hydrogen discharge at a cell temperature of 664 °C and 1 torr is shown in Figure 18. The breakdown voltage was approximately 220 V. The field voltage required to form a stable discharge was 224 V. The input power was 24.6 W. Spectral features are tabulated in Table III. The peak at 589.1 nm may be due to sodium contamination from a previous experimental run. The minor peaks at 518.2 and 558.7 nm have not been identified.

The spectrometer system irradiation for mixtures of hydrogen and sodium vapor are shown in Figures 19-21 for temperatures of 335, 516, and 664 °C, respectively. Corresponding hydrogen pressures are 1, 1.5, and 1.5 torr, respectively. The calculated sodium vapor pressure was 51 mtorr, 5.3 torr, and 63 torr at 335, 516, and 664 °C, respectively. At least 200 V was required to maintain a discharge. The input power for a stable discharge ranged from approximately 10 W at 664 °C to 34 W at 335 °C. Spectral features corresponding to 335 °C are summarized in Table IV. Strong emission observed near 656-657 nm was probably due, in-part, to hydrogen. The relative contribution to the intensity was masked by strong sodium emission at a slightly shorter wavelength. The peak at 486.2 nm could only be due to hydrogen emission. Sodium does not have emission lines in the neighborhood of this wavelength. The intensity of this peak diminishes relative to the more prominent sodium peaks with increasing temperature as shown in Figures 19-21. This may have been due to a decreasing hydrogen concentration as the sodium vapor pressure increased.

The spectral response for mixtures of magnesium vapor and hydrogen are shown in Figures 22-24 for temperatures of 449, 582, and 654 °C, respectively. The corresponding hydrogen pressures are 4, 4.2, and 3 torr, respectively. A minimum of 150 V was required to maintain a discharge. The minimum input power required to maintain a stable discharge was 58 W at 449 °C. Spectral features corresponding to 449 °C are summarized in Table V. Both hydrogen and magnesium spectral features are observed. The modest sodium emission at 588 nm may be due to sodium contamination from previous control experiments.

The spectral response for a mixture of barium vapor and hydrogen at 666°C is shown in Figure 25. The hydrogen partial pressure and barium vapor pressure are 2 torr and 25 mtorr, respectively. It was not possible to achieve a total visible irradiation level of $1\,\mu W/cm^2$ even with voltages approaching 150 V. The voltage and power input corresponding to Figure 25 are 138 V and 55 W, respectively. Spectral features are summarized in Table VI. Both barium and hydrogen spectral features are observed as well as sodium features which are presumably due to contamination. The peak at 493 nm has not been identified.

The spectral response for a mixture of hydrogen, argon, and strontium vapor at 514°C is shown in Figure 26. The hydrogen and argon partial pressures were 0.3 and 1 Torr, respectively. The strontium vapor pressure was approximately 6 mtorr at this temperature. The voltage required for a stable discharge was 6.6 V. The corresponding power input to the cell was approximately 4 mW. The spectral characteristics are tabulated in Table VII. The two major features were due to strontium emission. The very minor spectral features at 639 and 654 nm were due to argon. The total visible irradiation of the spectrometer detector was 1.3 μW/cm^2. The spectral responses for a mixture of argon and hydrogen and for pure argon are shown in Figures 27 and 28, respectively. For irradiation levels in the range 1-2 μW/cm^2, voltages of 224 and 190 V and power inputs of 33.5 and 24.7 W were required for discharges in argon and hydrogen, and pure argon, respectively. The spectral characteristics are tabulated in Tables VIII and IX for these gas discharges. In the case of pure argon, the emission at 656 nm was most likely hydrogen Balmer alpha emission due to some hydrogen contamination.

IV. DISCUSSION

Intense EUV emission was observed at low temperatures (e.g. $\approx 10^3\,K$) from atomic hydrogen and strontium which ionizes at integer multiple of the potential energy of atomic hydrogen. The emission intensity of the plasma generated by atomic strontium increased significantly with the introduction of argon gas only when Ar^+ emission was observed. The ionization energy of Ar^+ to Ar^{2+} is 27.6 eV. In the

cases where Lyman α emission was observed, no possible chemical reactions of the tungsten filament, the dissociator, the vaporized strontium, and 300 mtorr hydrogen or argon-hydrogen mixture at a cell temperature of 700 °C could be found which accounted for the hydrogen α line emission. In fact, no known chemical reaction releases enough energy to excite Lyman α emission from hydrogen. The emission was not observed with hydrogen or an argon-hydrogen mixture alone or with helium, neon, or argon gas. Intense emission was observed for strontium with hydrogen gas, but no emission was observed with hydrogen or strontium alone. This result indicates that the emission may be due to a reaction of hydrogen. The increase in intensity with the formation of Ar^+ and the equal dependency of the emission on the presence of both Ar^+ and atomic hydrogen indicates a reaction between these species.

Other studies support the possibility of a novel reaction of atomic hydrogen which produces an anomalous discharge. It has been previously reported that intense extreme ultraviolet (EUV) emission was observed at low temperatures (e.g. $\approx 10^3\,K$) from atomic hydrogen and certain atomized elements or certain gaseous ions [4-18]. The only pure elements that were observed to emit EUV were those wherein the ionization of t electrons from an atom to a continuum energy level is such that the sum of the ionization energies of the t electrons is approximately $m \cdot 27.2\,eV$ where t and m are each an integer. Strontium atoms ionize at integer multiples of the potential energy of atomic hydrogen and caused emission. Whereas, the chemically similar atoms, sodium, magnesium and barium, do not ionize at integer multiples of the potential energy of atomic hydrogen and caused no emission. The ionization of Sr to Sr^{5+} has a net enthalpy of reaction of $188.2\,eV$ [19], which is equivalent to $m = 7$. Argon ions can also provide a net enthalpy of a multiple of that of the potential energy of the hydrogen atom. The second ionization energy of argon is $27.63\,eV$. The reaction Ar^+ to Ar^{2+} has a net enthalpy of reaction of $27.63\,eV$, which is equivalent to $m = 1$.

The power balance of the gas cell having atomized hydrogen and strontium was measured by integrating the total light output corrected for spectrometer system response and energy over the visible range. A control cell was identical except that sodium, magnesium, or barium replaced strontium. In the hydrogen controls, 4000-7000 times the

power of the strontium cell was required in order to achieve the same optically measured light output power. With the addition of argon to the hydrogen-strontium plasma, the light output for power input increased by a factor of about two. In the argon-hydrogen controls up to 8600 times the power input was required to match the light output of the argon-hydrogen-strontium plasma. A plasma formed at a cell voltage of about 250 V for hydrogen alone and sodium-hydrogen mixtures, 140-150 V for hydrogen-magnesium and hydrogen-barium mixtures, 224 V for an argon-hydrogen mixture alone, and 190 V for argon alone; whereas, a plasma formed for hydrogen-strontium mixtures and argon-hydrogen-strontium mixtures at extremely low voltages of about 2 V and 6.6 V, respectively. This is two orders of magnitude lower than the starting voltages measured for gas glow discharges, cf. Table X.

An anomalous plasma with hydrogen-potassium mixtures has been reported in an experiment identical to the present EUV experiments [9-10, 12]. In experiments performed at the Institut Fur Niedertemperatur-Plasmaphysik e.V., an anomalous plasma formed with hydrogen/potassium mixtures wherein the plasma decayed with a two second half-life which was the thermal decay time of the filament which dissociated molecular hydrogen to atomic hydrogen when the electric field was set to zero [9, 12]. This experiment showed that hydrogen line emission was occurring even though the voltage between the heater wires was set to and measured to be zero and indicated that the emission was due to a reaction of potassium atoms with atomic hydrogen. Potassium atoms ionize at an integer multiple of the potential energy of atomic hydrogen, $m \cdot 27.2 \, eV$. The enthalpy of ionization of K to K^{3+} has a net enthalpy of reaction of $81.7426 \, eV$, which is equivalent to $m = 3$.

An anomalous plasma of hydrogen and certain alkali ions formed at low temperatures (e.g. $\approx 10^3 \, K$) as recorded via EUV spectroscopy and the hydrogen Balmer and alkali line emissions in the visible range [10]. The observed plasma formed at low temperatures (e.g. $\approx 10^3 \, K$) from atomic hydrogen generated at a tungsten filament that heated a titanium dissociator and one of potassium, rubidium, cesium, and their carbonates and nitrates. These atoms and ions ionize to provide a net enthalpy of reaction of an integer multiple of the potential energy of atomic hydrogen ($m \cdot 27.2 \, eV$, $m = \text{integer}$) to within 0.17 eV and comprise only a single

ionization in the case of a potassium or rubidium ion. Whereas, the chemically similar atoms of sodium and sodium and lithium carbonates and nitrates which do not ionize with these constraints caused no emission. To test the electric dependence of the emission, the weak electric field of about 1 V/cm was set and measured to be zero in $< 0.5 \times 10^{-6}$ sec. An anomalous afterglow duration of about one to two seconds was recorded in the case of potassium, rubidium, cesium, K_2CO_3, $RbNO_3$, and $CsNO_3$. Hydrogen line or alkali line emission was occurring even though the voltage between the heater wires was set to and measured to be zero. These atoms and ions ionize to provide a net enthalpy of reaction of an integer multiple of the potential energy of atomic hydrogen to within less than the thermal energies at $\approx 10^3 K$ and comprise only a single ionization in the case of a potassium or rubidium ion. Since the thermal decay time of the filament for dissociation of molecular hydrogen to atomic hydrogen was similar to the anomalous plasma afterglow duration, the emission was determined to be due to a reaction of atomic hydrogen with each of the atoms or ions that did not require the presence of an electric field to be functional.

In the present experiments, it was determined that the presence of a weak electric field was necessary in order for strontium to produce an anomalous discharge of hydrogen. In the case that electrons are ionized to a continuum energy level, the presence of a low strength electric field alters the continuum energy levels. For strontium, the minimum electric field in this experiment was about 2 V over the annular gap of about 2 cm. The ionization energy of $188.2\ eV$ is 1% less than $m \cdot 27.2\ eV$ where $m = 7$. In the anomalous discharge of hydrogen due to the presence of strontium, the weak field may adjust the energy of ionizing strontium to match the energy of $m \cdot 27.2\ eV$ to permit a novel reaction of atomic hydrogen. For Ar^+, the minimum electric field in this experiment was about 6.6 V. The ionization energy of Ar^+ to Ar^{2+} is $27.6\ eV$, and an electric field may adjust the energy of ionizing Ar^+ to Ar^{2+} to match the energy of $27.2\ eV$.

Reports of the formation of novel compounds provide substantial evidence supporting a novel reaction of hydrogen as the mechanism of the observed EUV emission and anomalous discharge. Novel hydrogen compounds have been isolated as products of the reaction of atomic

hydrogen with atoms and ions which formed an anomalous plasma as reported in the EUV studies [4-18, 25-39]. Novel inorganic alkali and alkaline earth hydrides of the formula MH^* and MH^*X wherein M is the metal, X, is a singly negatively charged anion, and H^* comprises a novel high binding energy hydride ion were synthesized in a high temperature gas cell by reaction of atomic hydrogen with a catalyst such as potassium metal and MH, MX or MX_2 corresponding to an alkali metal or alkaline earth metal compound, respectively [25-26, 29]. Novel hydride compounds were identified by 1.) time of flight secondary ion mass spectroscopy which showed a dominant hydride ion in the negative ion spectrum, 2.) X-ray photoelectron spectroscopy which showed novel hydride peaks and significant shifts of the core levels of the primary elements bound to the novel hydride ions, 3.) ^1H nuclear magnetic resonance spectroscopy (NMR) which showed extraordinary upfield chemical shifts compared to the NMR of the corresponding ordinary hydrides, and 4.) thermal decomposition with analysis by gas chromatography, and mass spectroscopy which identified the compounds as hydrides [26, 29]. The implications are that a new field of novel hydrogen chemistry has been discovered. The catalytic reaction of hydrogen may give rise to an energetic anomalous discharge such as an anomalous strontium-argon-hydrogen discharge.

ACKNOWLEDGMENT

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Table I. Discharge conditions and comparison of the driving power to achieve a total visible radiant flux of about $1\,\mu W/cm^2$.

	T (°C)	$P_{hyd.}$ (torr)	P_{Ar} (torr)	P_v (torr)[a]	Voltage (V)	Current (mA)	Integ. time (ms)	Detector irradiation ($\mu W/cm^2$)	Power (W)
Ar+H$_2$+Sr	514	0.3	1.0	0.006	6.56	0.6	204	1.3	0.0039
Ar+H$_2$	519	0.295	0.5	- - - -	224	184	409	1.9	33.5[b]
Ar	520	- - - -	1.0	- - - -	190	170	307	1.1	24.7[b]
H$_2$+Sr	664	- - - -		0.270	2.20	3.86	768	1.17	0.0085
H$_2$	664	1.0		- - - -	224	110	1130	2.08	24.6
H$_2$+Na	335	1.0		0.051	272	124	122	1.85	33.7
H$_2$+Na	516	1.5		5.3	220	68	768	0.40	15.0
H$_2$+Na	664	1.5		63	240	41	768	0.41	9.84
H$_2$+Mg	449	4.0		0.016	153	380	500	1.7	58
H$_2$+Mg	582	4.2		0.6	233	290	500	0.16	68
H$_2$+Mg	654	3.0		2.8	250	400	1000	0.18	100.0
H$_2$+Ba	666	2.0		0.025	138	730	716	0.03	55[b]
Bkgnd.	664	- - - -		0.270	0	0	768	0.20	0

[a] Calculated [21]

[b] Power input differs from volt-amperes due to non-unity power factor.

TABLE II. Spectral features of hydrogen and strontium at 664 °C.

Measured Wavelength (*nm*)	Spectrometer System Irradiation $\left(\mu W/cm^2 nm\right)$	Published Emission Data [22] (*nm*)
460.6	0.156	460.73 (Sr)
487.2	0.00290	487.25 (Sr), 486.13 (H$_2$)
639.8	0.00813	638.82 (Sr)
654.7	0.0139	654.68 (Sr), 656.29 (H$_2$)
689.4	0.0386	689.26 (Sr)

TABLE III. Spectral features of hydrogen at 664 °C.

Measured Wavelength (nm)	Spectrometer System Irradiation $(\mu W / cm^2 nm)$	Published Emission Data [22] (nm)
485.8	0.0165	486.13 (H_2)
518.2	0.00894	
558.7	0.00694	
589.1	0.0174	589.00 (Na), 589.59 (Na)
656.7	0.0752	656.29 (H_2)

TABLE IV. Spectral features of hydrogen and sodium at 335 °C.

Measured wavelength (nm)	Spectrometer System Irradiation $(\mu W / cm^2 nm)$	Published emission data [22] (nm)
467.2	0.00400	466.86 (Na)
486.2	0.0055	486.13 (H_2)
498.4	0.0176	498.28 (Na)
516.1	0.00380	515.34 (Na)
569.0	0.114	568.82 (Na)
589.3	0.302	589.00 (Na), 589.59 (Na)
615.9	0.0310	616.07 (Na)
656.0	0.0422	656.29 (H_2), 655.24 (Na)
657.0	0.0421	656.29 (H_2)

TABLE V. Spectral features of hydrogen and magnesium at 449 °C.

Measured wavelength (nm)	Spectrometer system irradiation $(\mu W/cm^2 nm)$	Published emission data [22] (nm)
382.6	0.0843	382.93 (Mg), 383.23 (Mg)
384.0	0.0643	383.83 (Mg)
485.2	0.0122	486.13 (H_2)
517.3	0.0353	517.27 (Mg), 518.36 (Mg)
588.1	0.0167	589.00 (Na), 589.59 (Na)
655.8	0.109	656.29 (H_2)

Table VI. Spectral features of hydrogen and barium at 666°C.

Measured wavelength (nm)	Spectrometer system irradiation $(\mu W/cm^2 nm)$	Published emission data [22] (nm)
456.2	0.0021	455.40 (Ba)
492.6	0.002	
552.7	8.4×10^{-4}	553.55 (Ba)
568.4	0.003	568.26 (Na)
588.8	0.006	589.00 (Na)
614.7	9.0×10^{-4}	614.17 (Ba)
655.9	0.002	656.29 (H_2)

Table VII. Spectral features of argon, hydrogen, and strontium at 514 °C.

Measured wavelength (nm)	Spectrometer system irradiation $(\mu W/cm^2 nm)$	Published emission data [22] (nm)
459.9	0.176	460.73 (Sr)
639.4	0.004	638.47 (Ar)
653.9	0.007	653.81 (Ar)
688.8	0.116	687.84 (Sr)

Table VIII. Spectral features of argon and hydrogen at 519 °C.

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Measured wavelength (nm)	Spectrometer system irradiation $(\mu W/cm^2 nm)$	Published emission data [22] (nm)
486.0	0.032	486.13 (H$_2$)
588.5	0.020	588.86 (Ar)
656.2	0.176	656.29 (H$_2$)
738.5	0.034	738.40 (Ar)

Table IX.   Spectral features of argon at 520 °C.

Measured wavelength (nm)	Spectrometer system irradiation $(\mu W/cm^2 nm)$	Published emission data [22] (nm)
588.8	0.039	588.86 (Ar)
655.9	0.046	656.29 (H$_2$)
696.4	0.028	696.54 (Ar)
706.5	0.028	706.72 (Ar)
738.5	0.058	738.40 (Ar)

TABLE X.   Glow discharge parameters from von Engel [23] and Naidu Kamaraju [24].

Gas	Minimum starting voltage (V)	Pressure-discharge gap product at minimum starting voltage (cm-torr)
N$_2$	251	0.67
H$_2$	273	1.15
Air	327	0.567
CO$_2$	420	0.51
Ar	137	0.9
He	156	4.0
Hg	520	2
Na	335	0.04

Figure Captions

Figure 1. The experimental set up comprising a gas cell light source and an EUV spectrometer which was differentially pumped.

Figure 2. Cylindrical stainless steel gas cell for plasma studies with 1.) hydrogen, argon, or argon-hydrogen mixture alone, 2.) hydrogen with strontium, sodium, magnesium, or barium, and 3.) argon-hydrogen mixture (97/5%) with strontium.

Figure 3. The experimental setup for generating a glow discharge hydrogen plasma and for optically measuring the power balance.

Figure 4. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 5. The UV/VIS spectrum ($40-560\,nm$) of the cell emission from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, and 300 mtorr hydrogen that was recorded with a photomultiplier tube (PMT) and a sodium salicylate scintillator.

Figure 6. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized sodium metal, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 7. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized magnesium, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 8. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized barium metal, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 9. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized strontium metal, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 10. The EUV spectrum ($40-160\,nm$) of the cell emission

recorded at about the point of the maximum Lyman α emission from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, a titanium dissociator, vaporized strontium metal, and 300 mtorr hydrogen that was recorded with a CEM.

Figure 11. The intensity of the Lyman α emission as a function of time from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a CEM.

Figure 12. The EUV emission spectrum (40 – 160 nm) from a gas cell comprising a tungsten filament, vaporized sodium metal, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a PMT and a sodium salicylate scintillator two hours after the cell reached 700 °C.

Figure 13. The EUV emission spectrum (40 – 160 nm) from a gas cell comprising a tungsten filament, vaporized magnesium metal, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a PMT and a sodium salicylate scintillator two hours after the cell reached 700 °C.

Figure 14. The EUV emission spectrum (40 – 160 nm) from a gas cell comprising a tungsten filament, vaporized barium metal, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a PMT and a sodium salicylate scintillator two hours after the cell reached 700 °C.

Figure 15. The EUV spectrum (40 – 160 nm) of the cell emission recorded at about the point of the maximum Lyman α emission from the gas cell at a cell temperature of 700 °C comprising a tungsten filament, vaporized strontium metal, and 300 mtorr argon-hydrogen mixture (97/3%) that was recorded with a PMT and a sodium salicylate scintillator.

Figure 16. Count rate and spectrometer system irradiation of the background spectrum of hydrogen and strontium vapor over the wavelength range $350 \leq \lambda \leq 750$ nm in the absence of power applied to the electrode and in the absence of a discharge.

Figure 17. The count rate and the spectrometer system irradiation for a mixture of hydrogen and strontium vapor at 664 °C.

Figure 18. The spectrometer system irradiation for a hydrogen discharge at a cell temperature of 664 °C and a hydrogen pressure of 1 torr.

Figure 19. The spectrometer system irradiation for a mixture of hydrogen and sodium vapor at 335 °C.

Figure 20. The spectrometer system irradiation for a mixture of hydrogen and sodium vapor at 516 °C.

Figure 21. The spectrometer system irradiation for a mixture of hydrogen and sodium vapor at 664 °C.

Figure 22. The spectrometer system irradiation for a mixture of hydrogen and magnesium vapor at 449 °C.

Figure 23. The spectrometer system irradiation for a mixture of hydrogen and magnesium vapor at 582 °C.

Figure 24. The spectrometer system irradiation for a mixture of hydrogen and magnesium vapor at 654 °C.

Figure 25. The spectrometer system irradiation for a mixture of hydrogen and barium vapor at 666 °C.

Figure 26. The spectrometer system irradiation for a mixture of argon-hydrogen and strontium vapor at 514 °C.

Figure 27. The spectrometer system irradiation for a mixture of argon and hydrogen at 519 °C.

Figure 28. The spectrometer system irradiation for an argon discharge at a cell temperature of 520 °C and argon pressure of 1 torr.
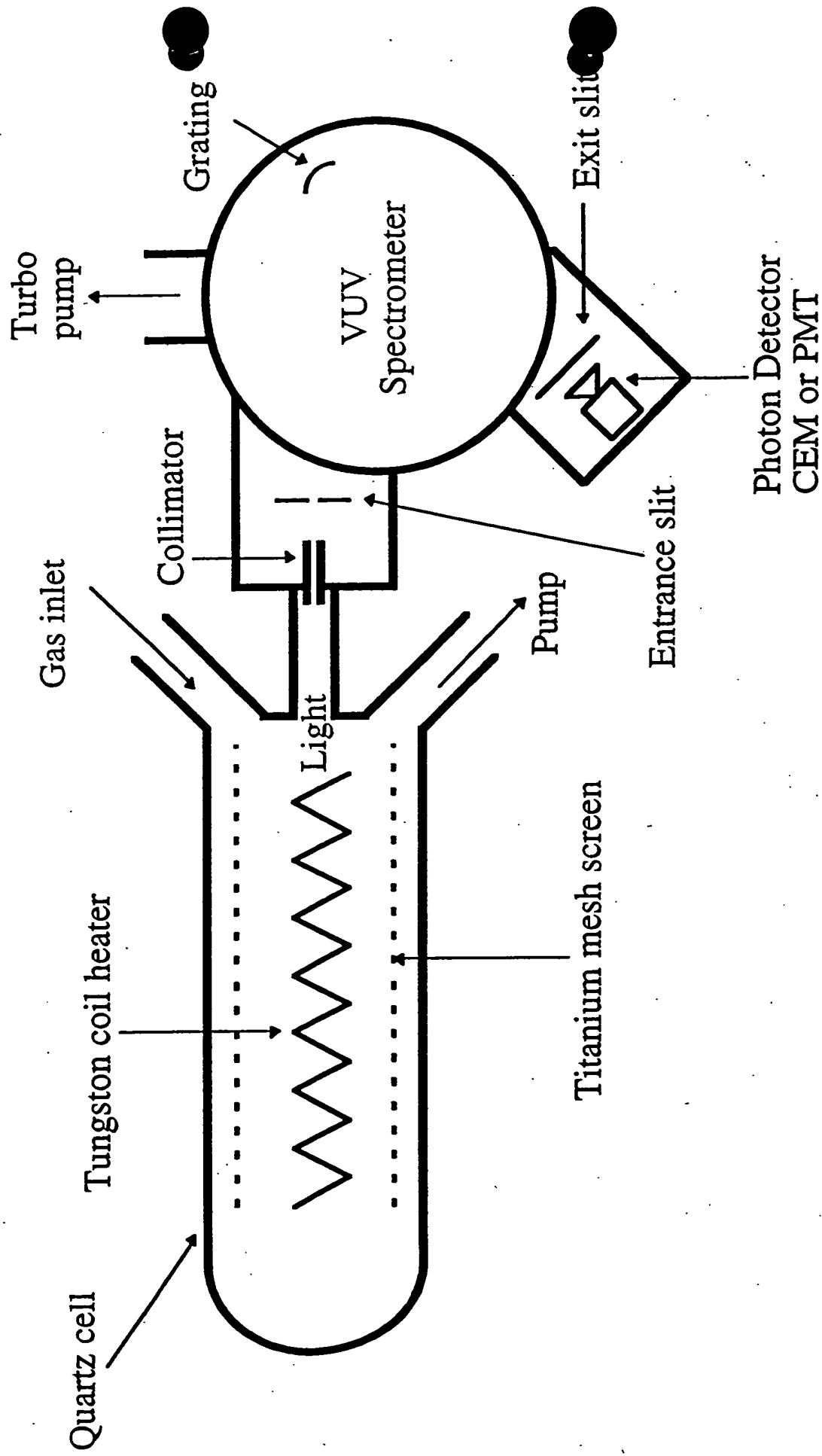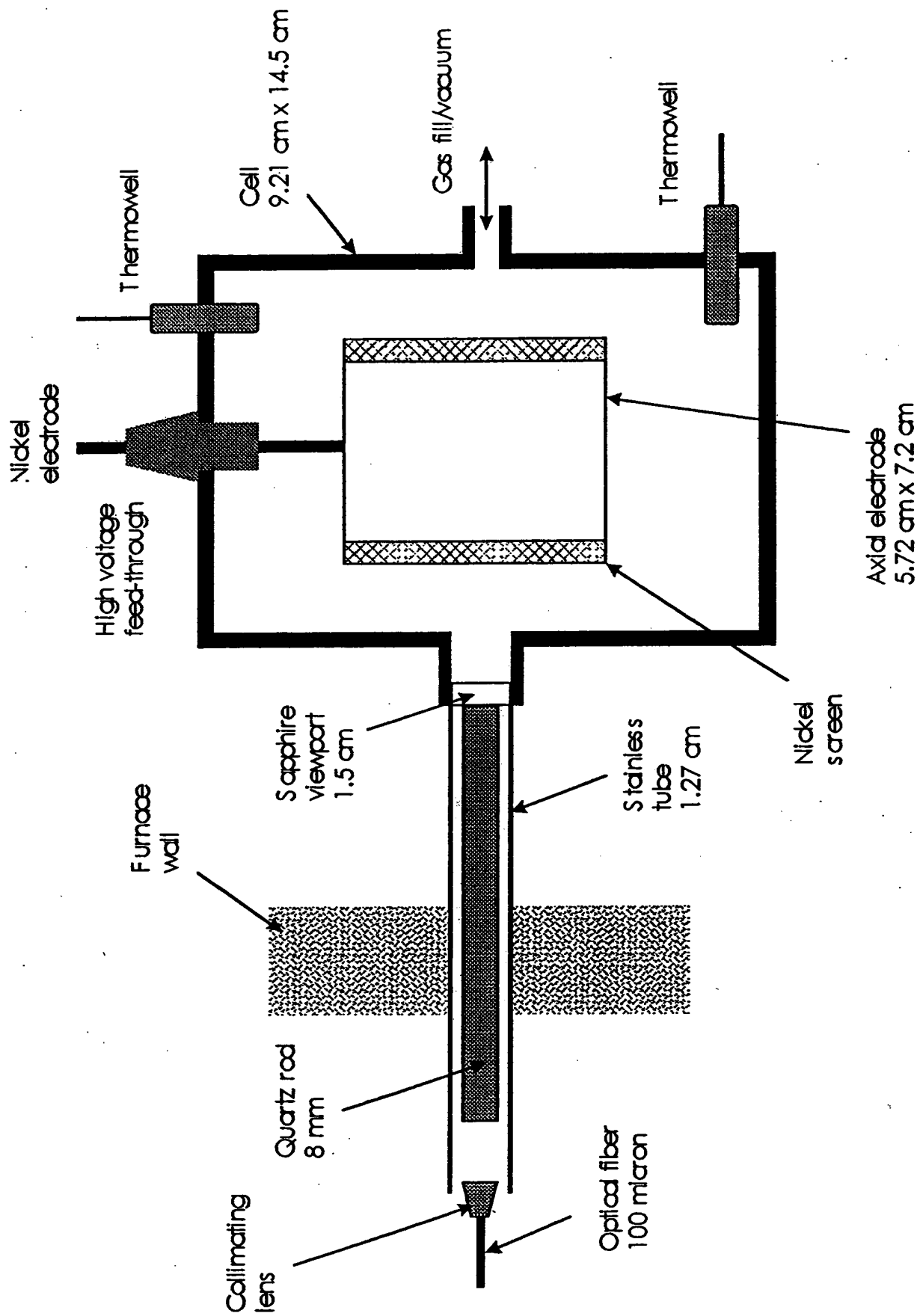
Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Hydrogen gas only

Photon Counts/sec

Wavelength (nm)
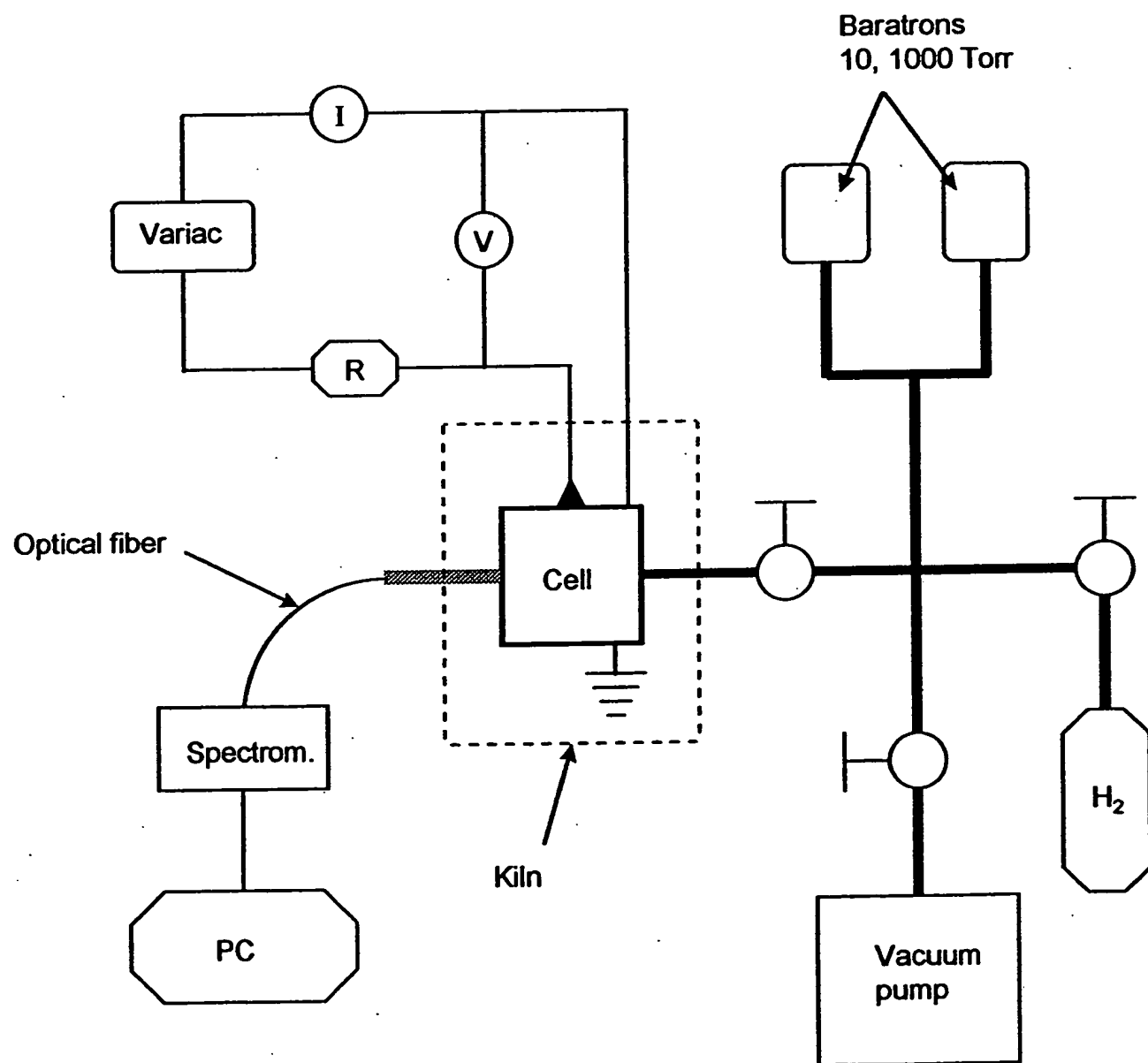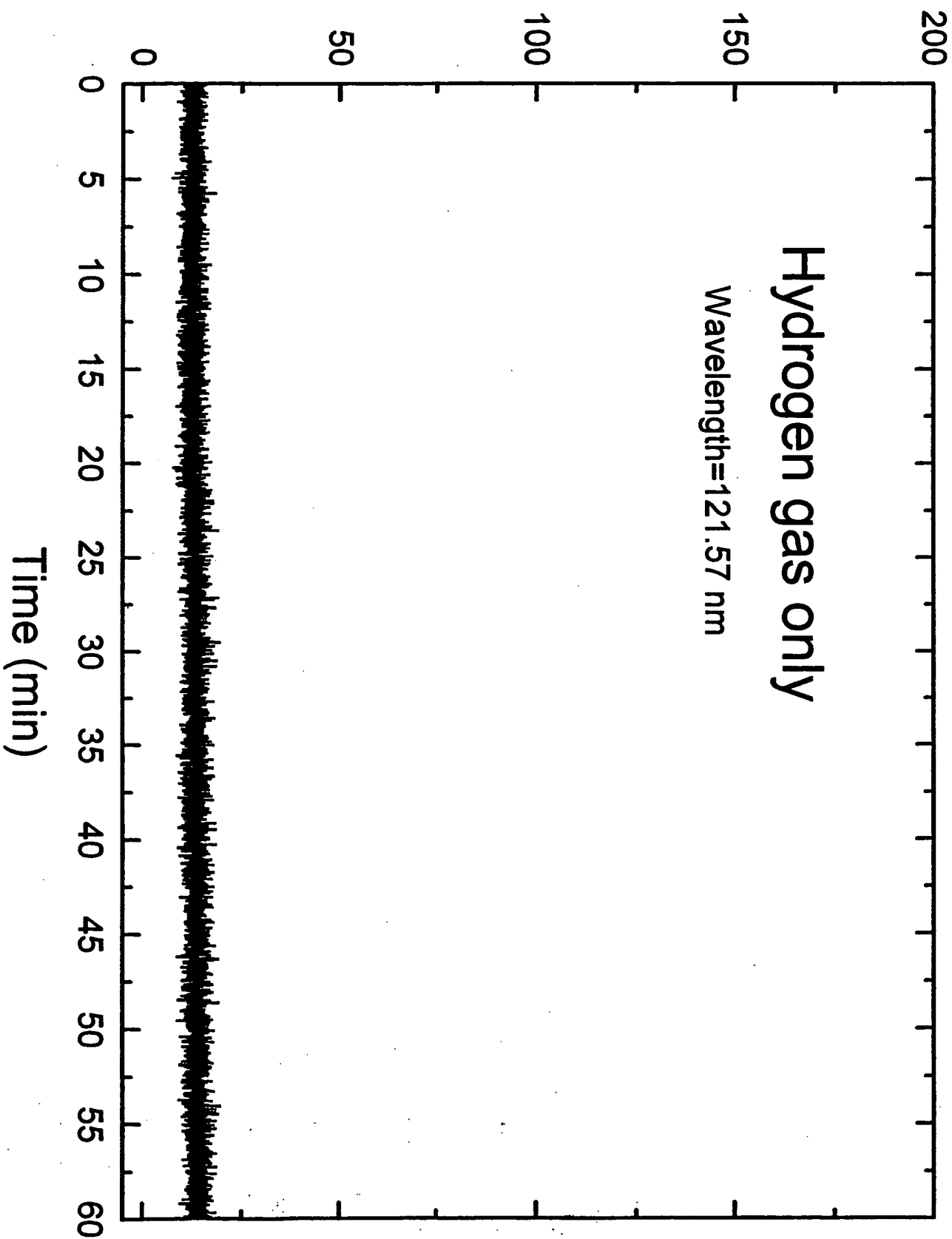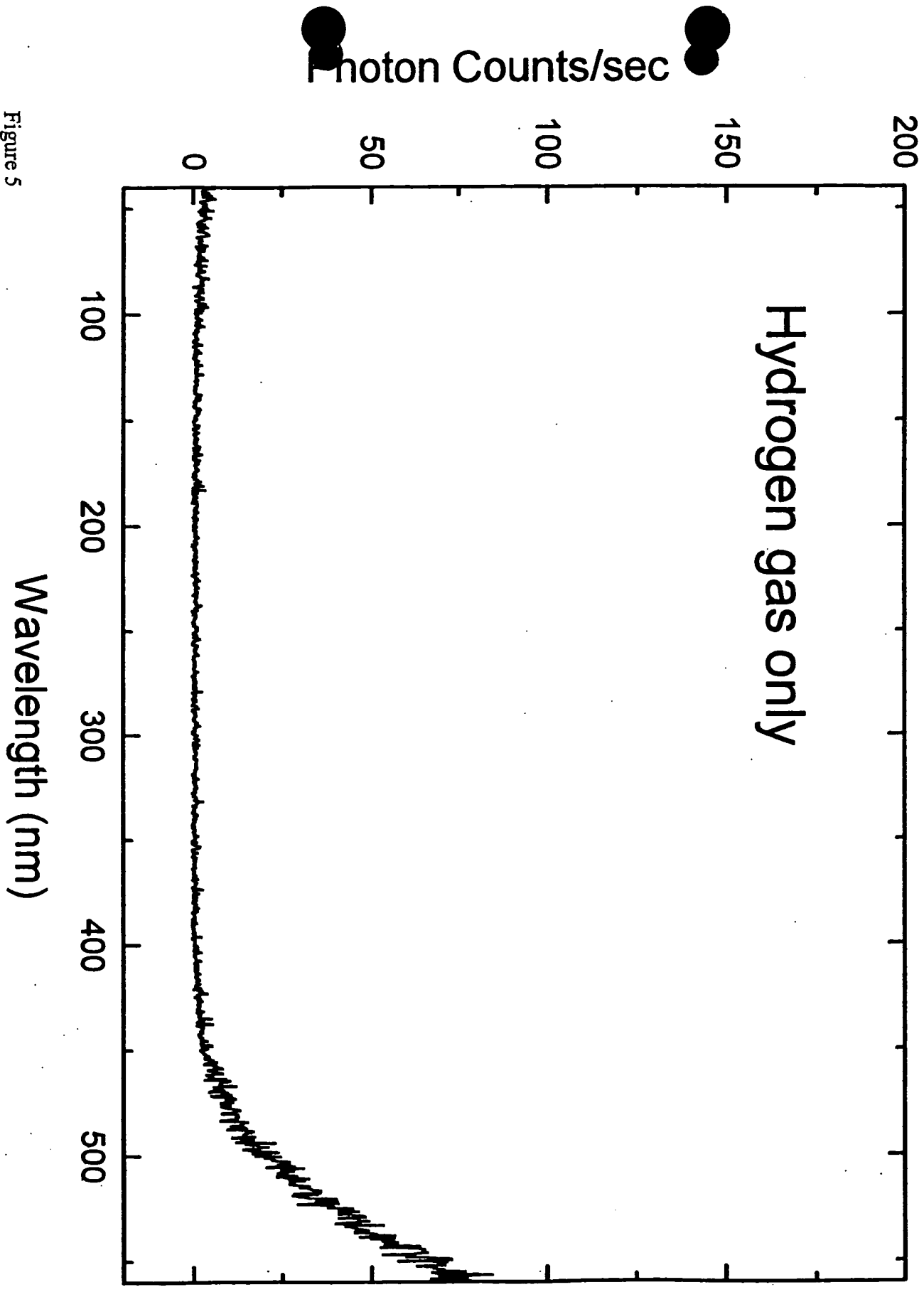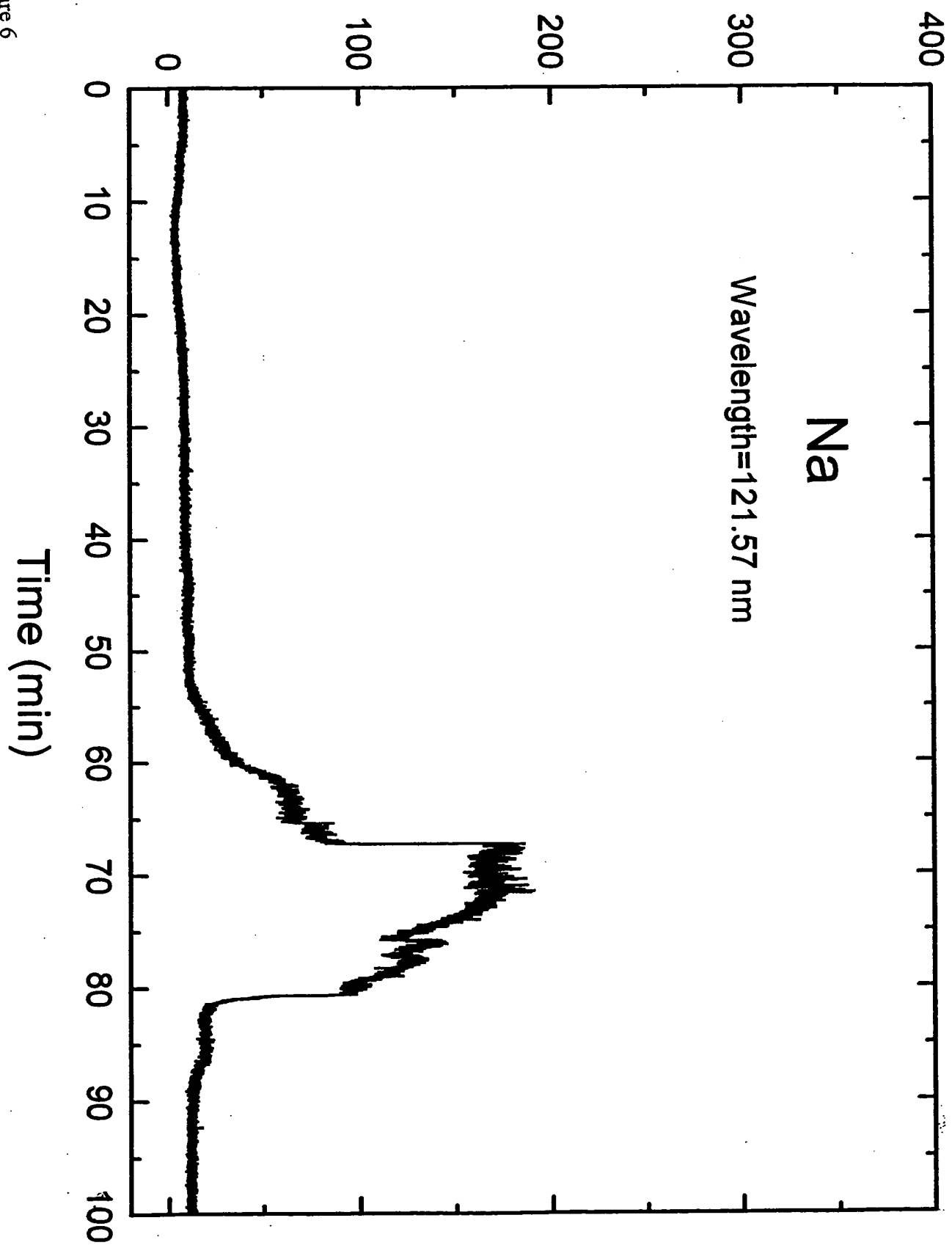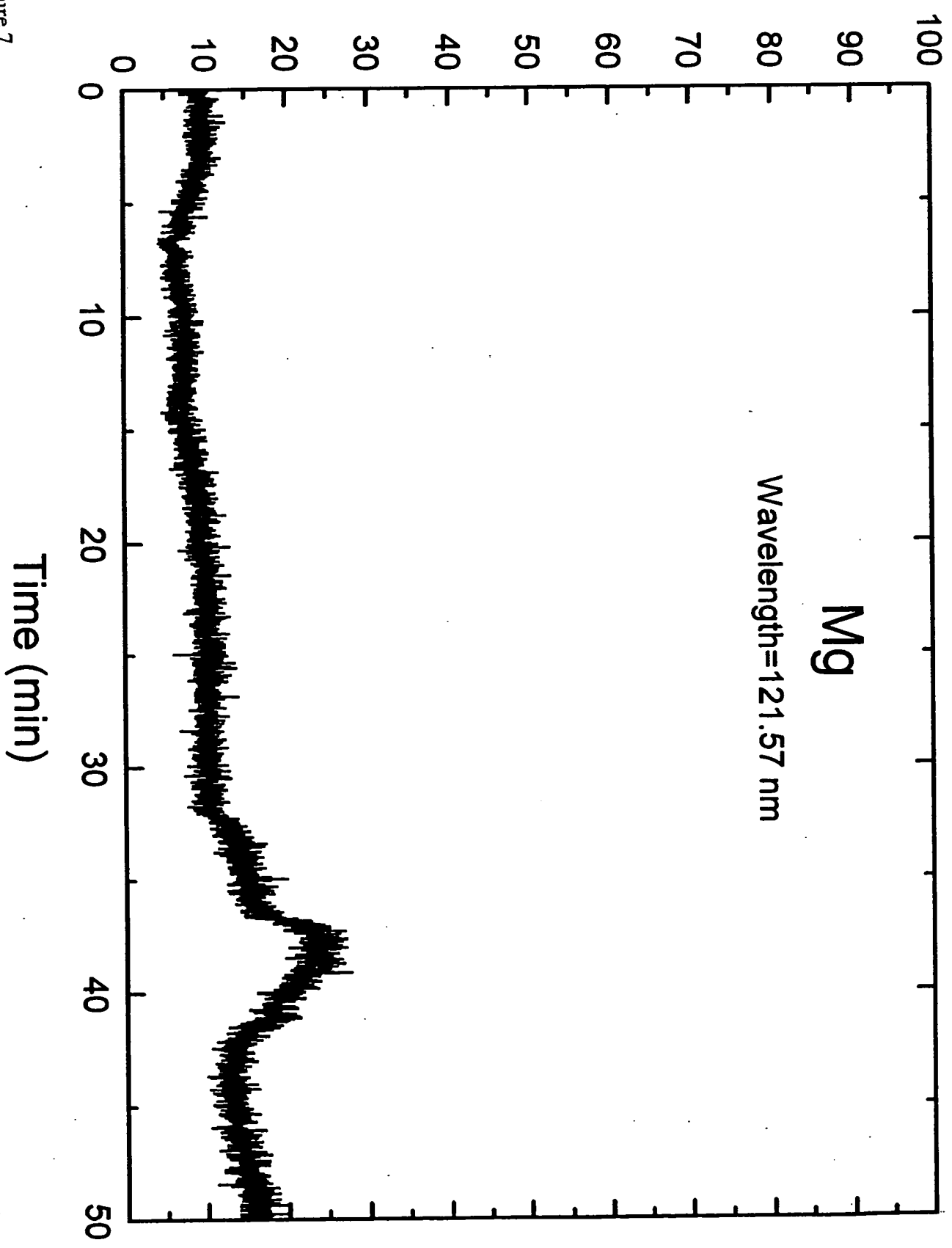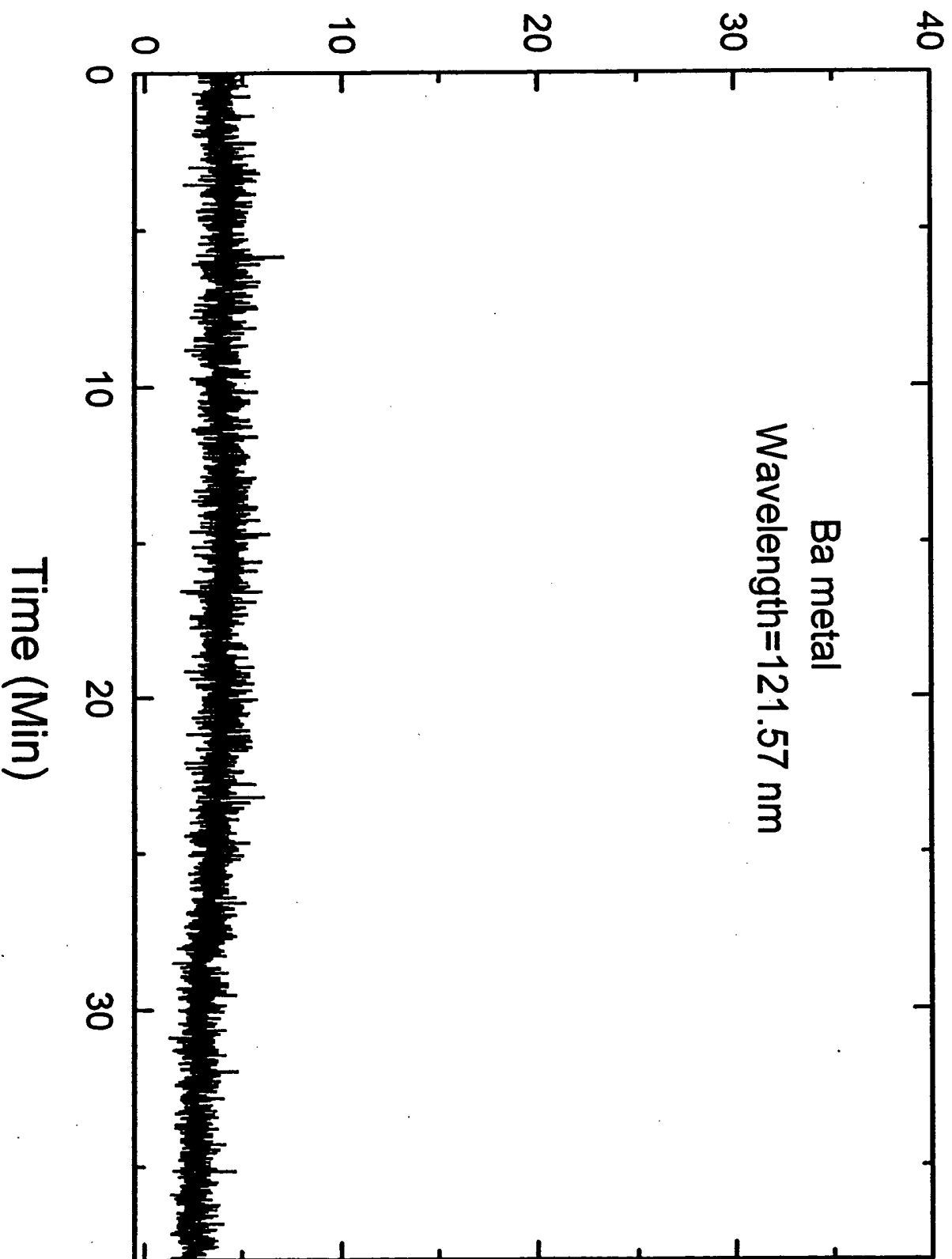
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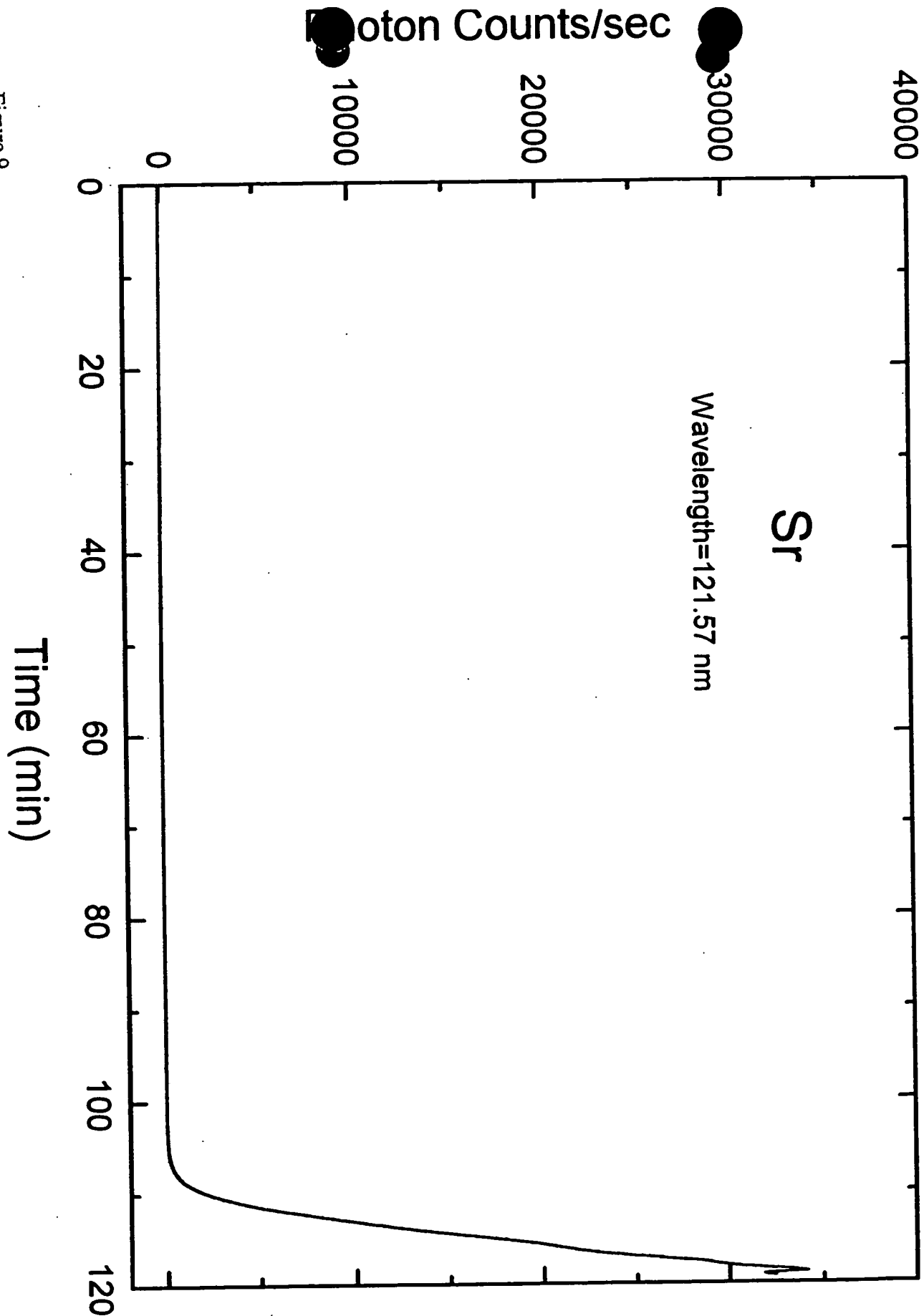
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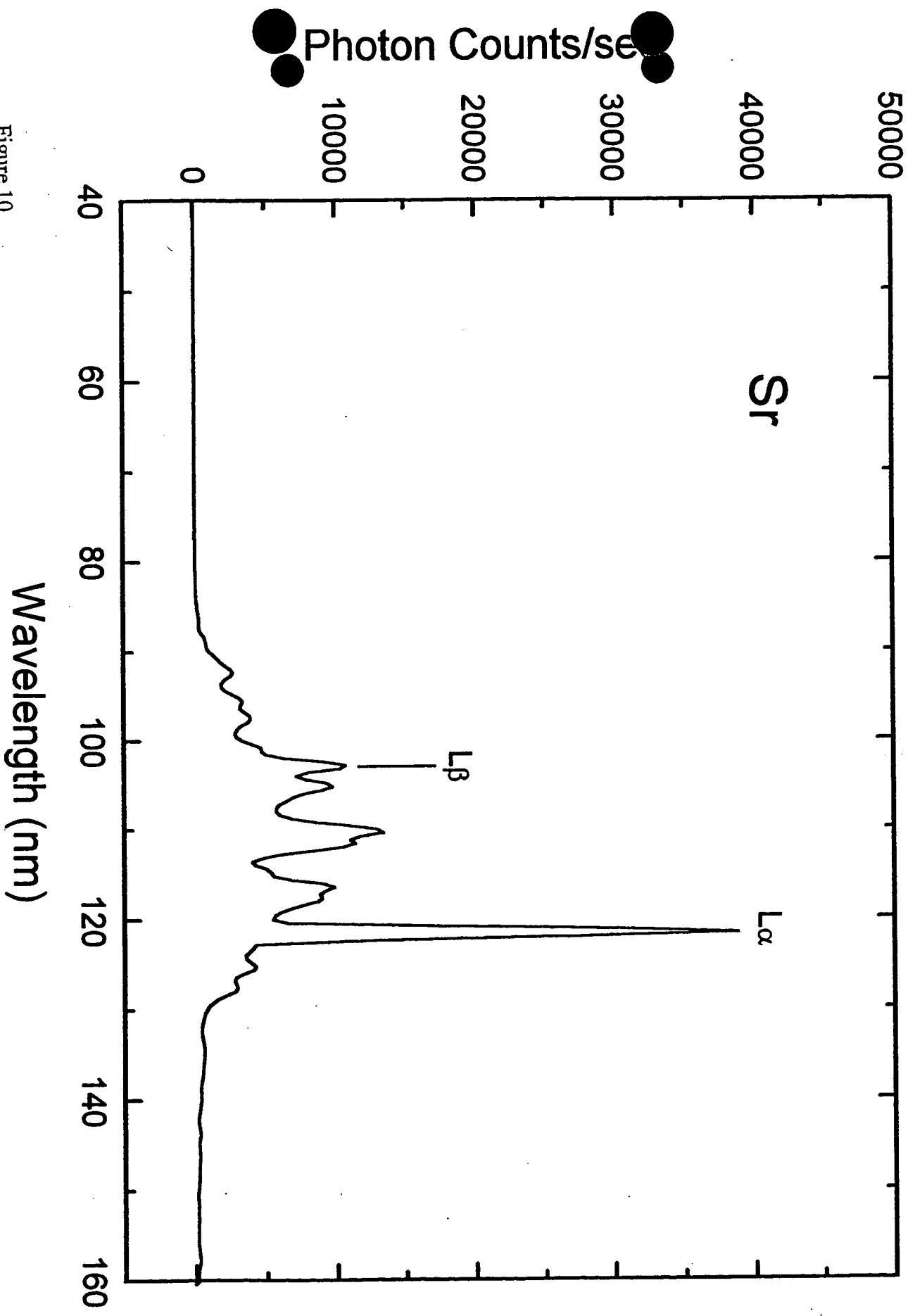
Figure 8

Figure 9

Figure 10

Figure 11

Na/Ar, H_2

Wavelength (nm)
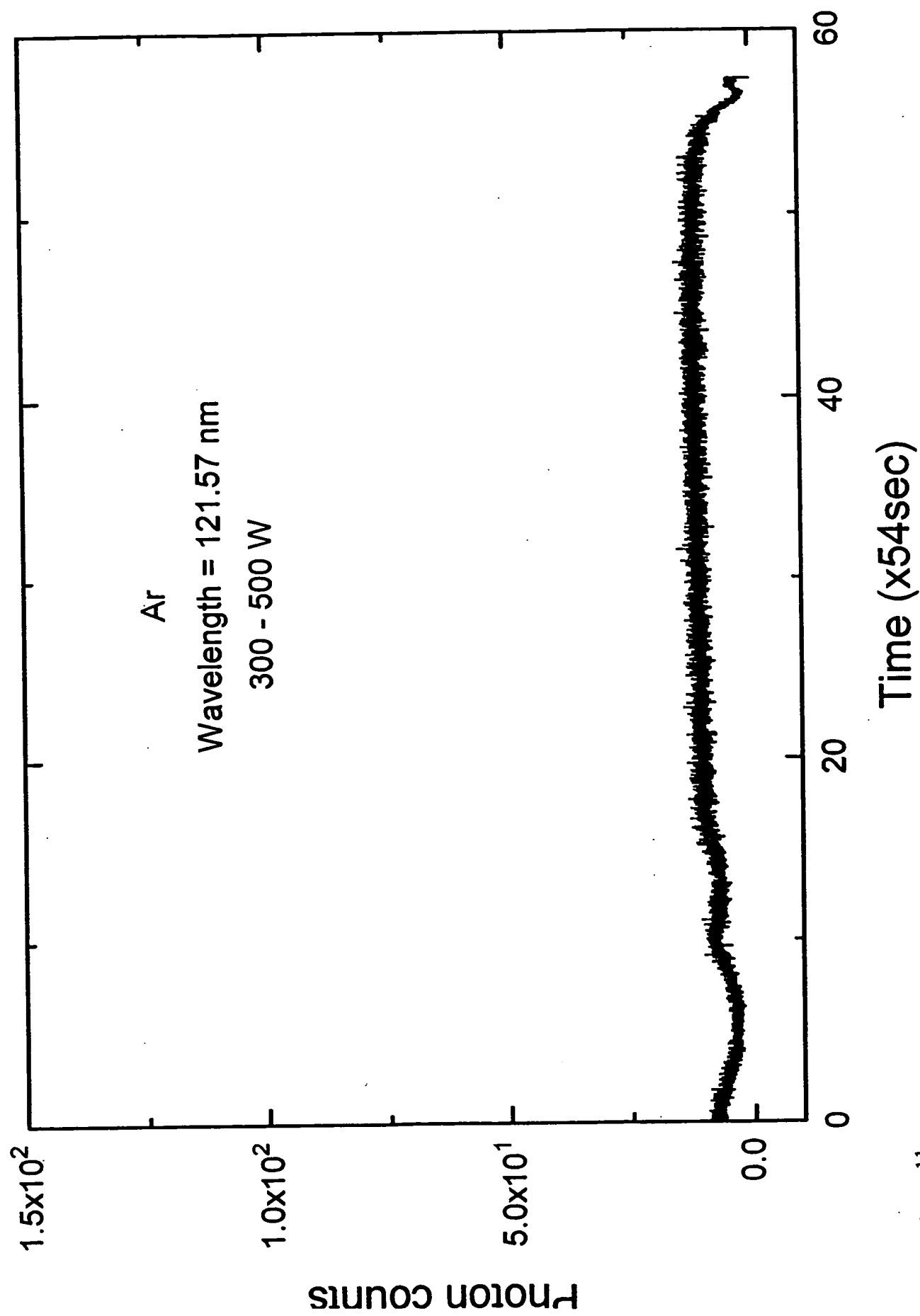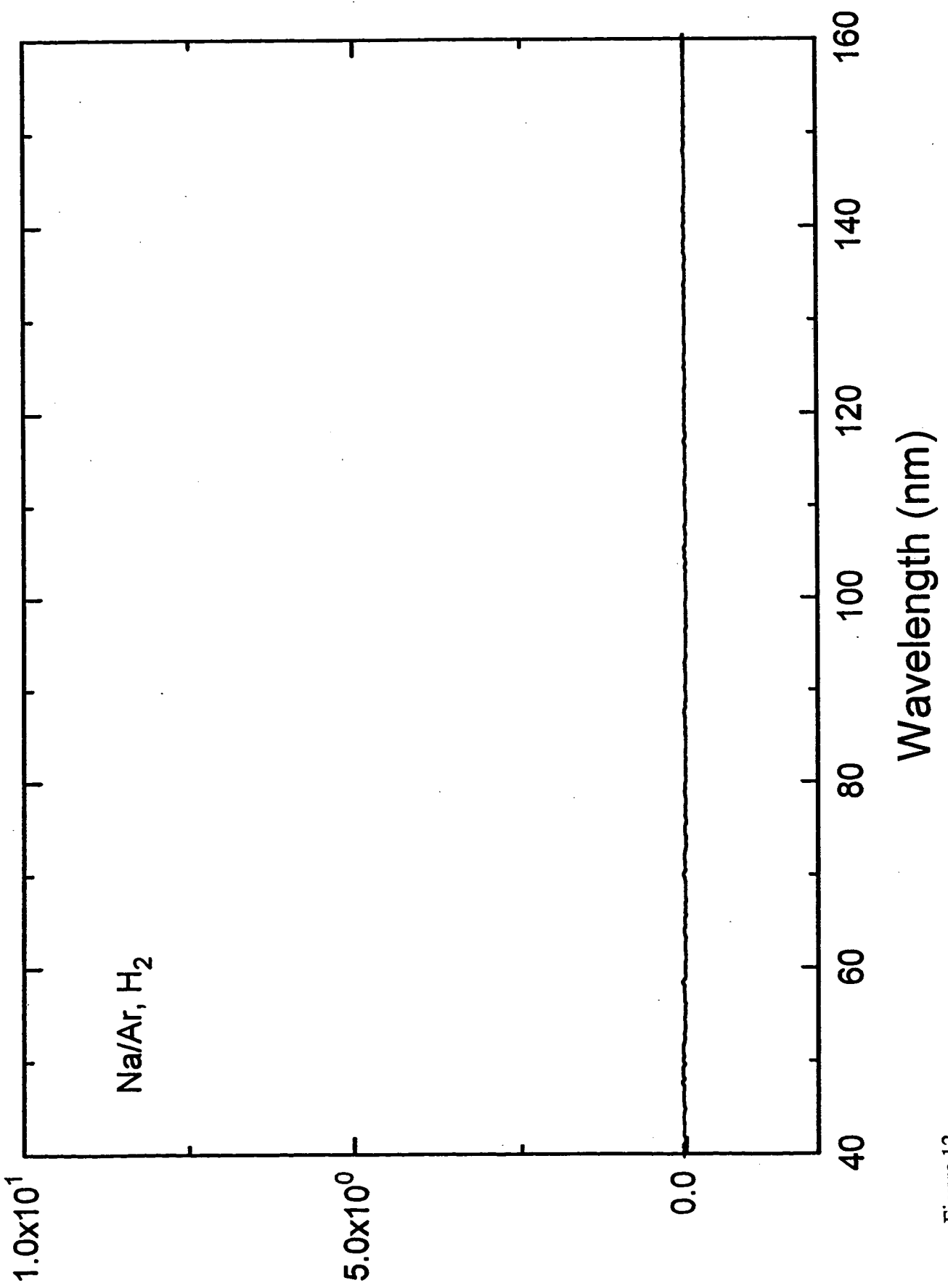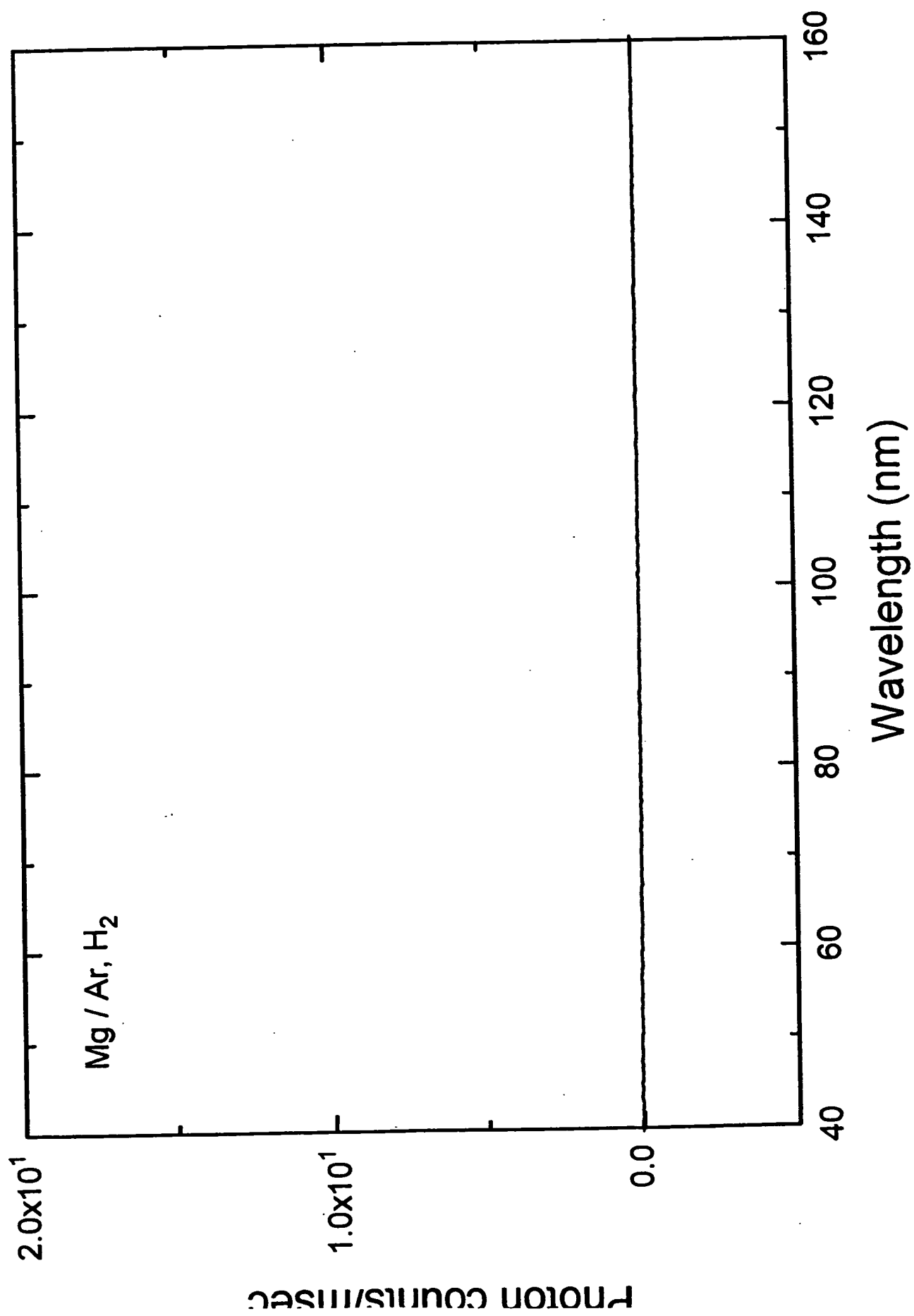
Figure 12

Figure 13
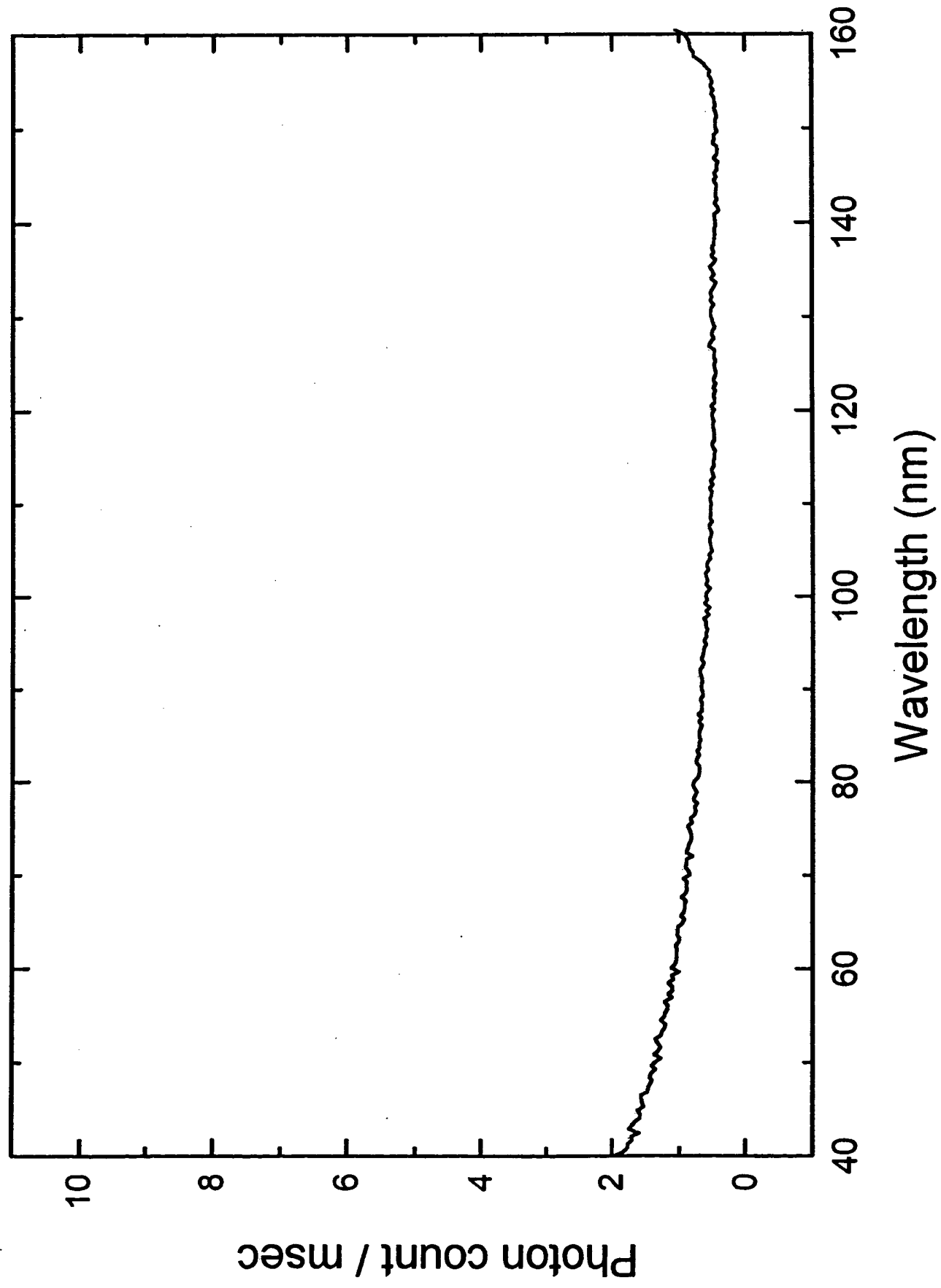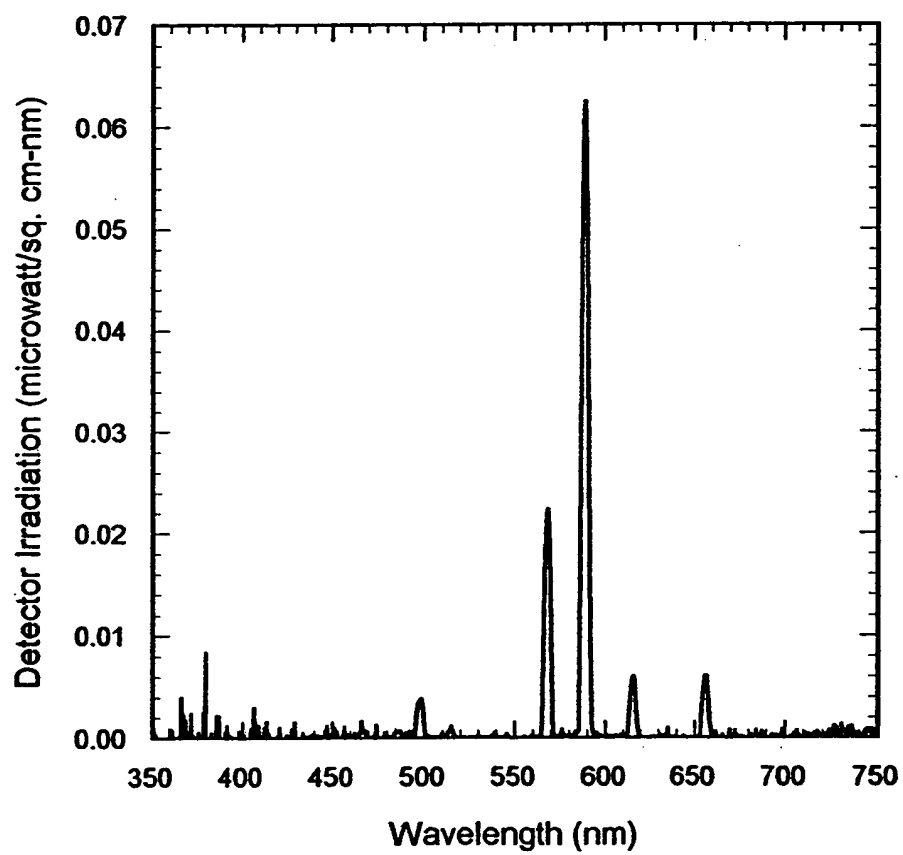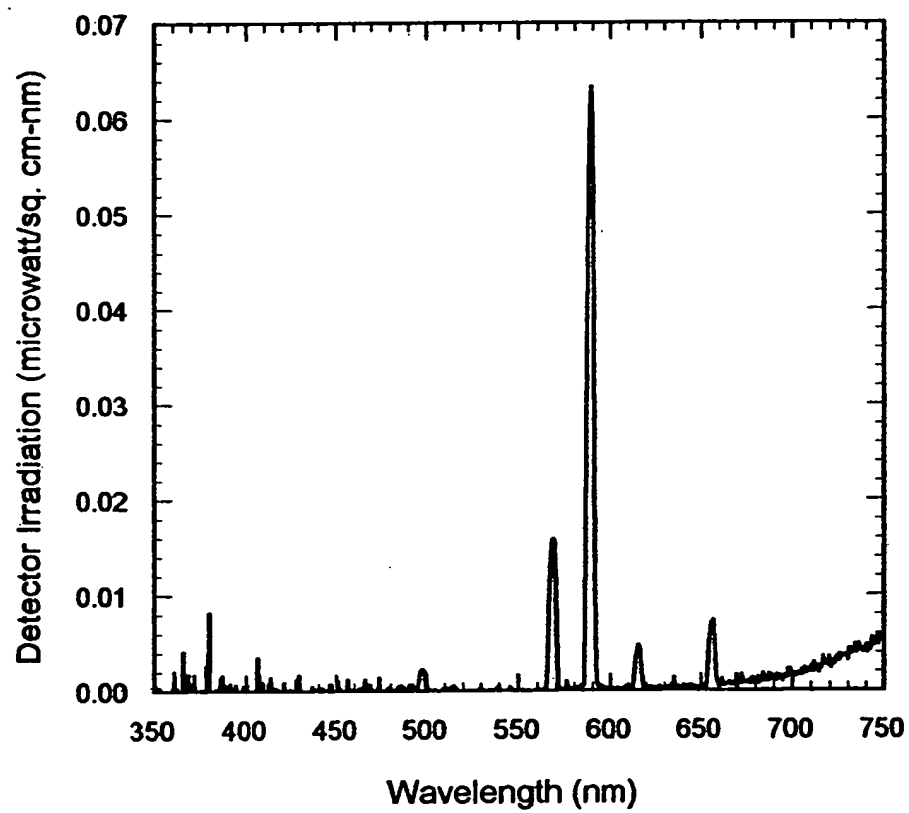
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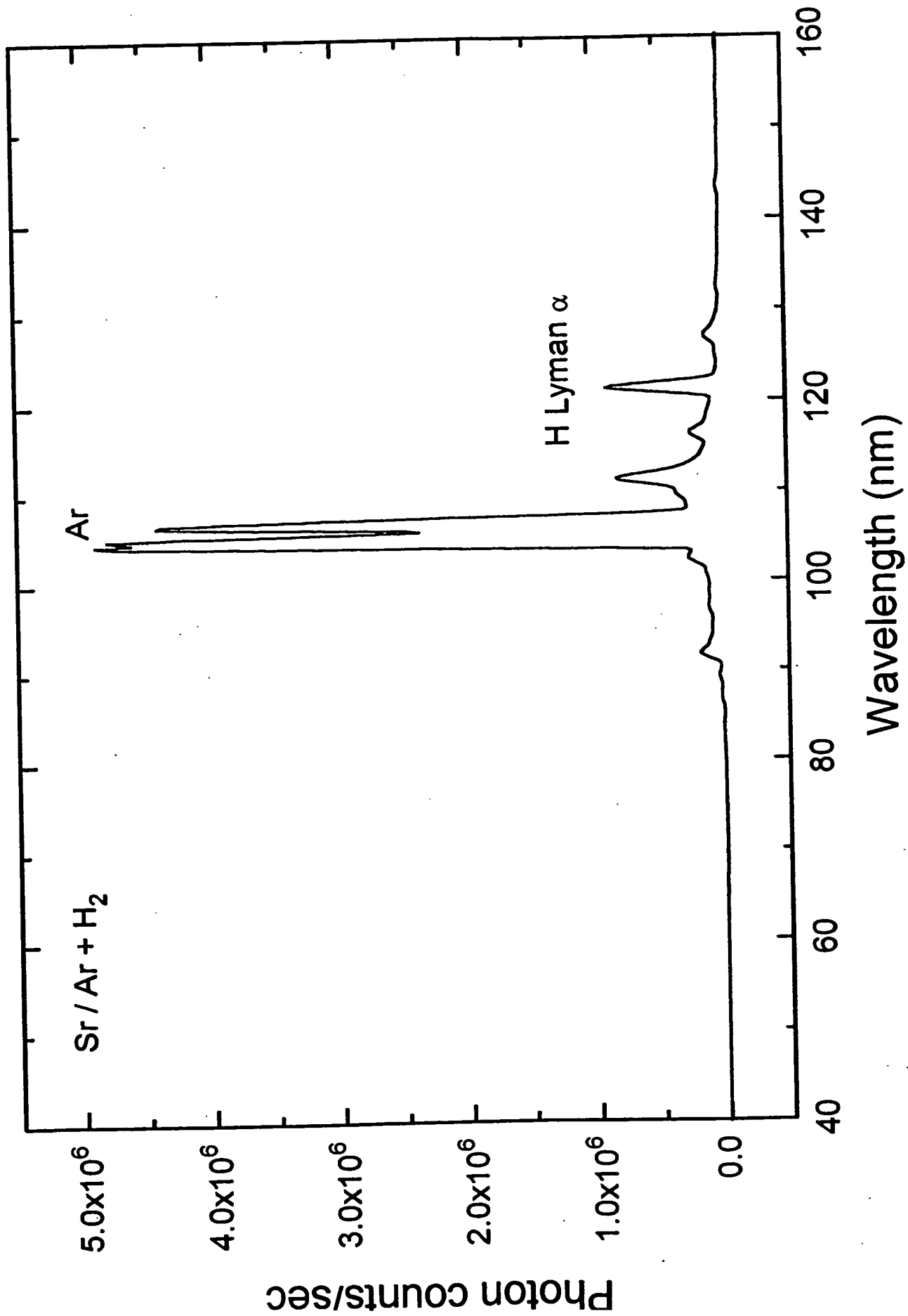
Figure 15

Figure 16
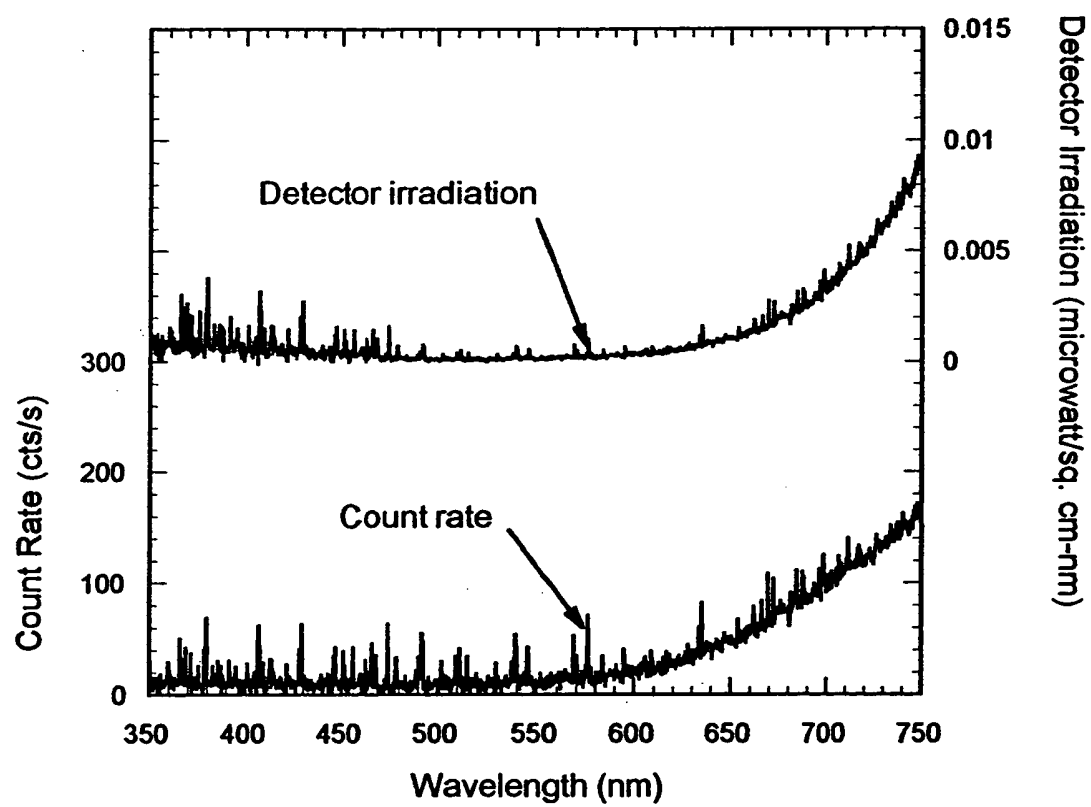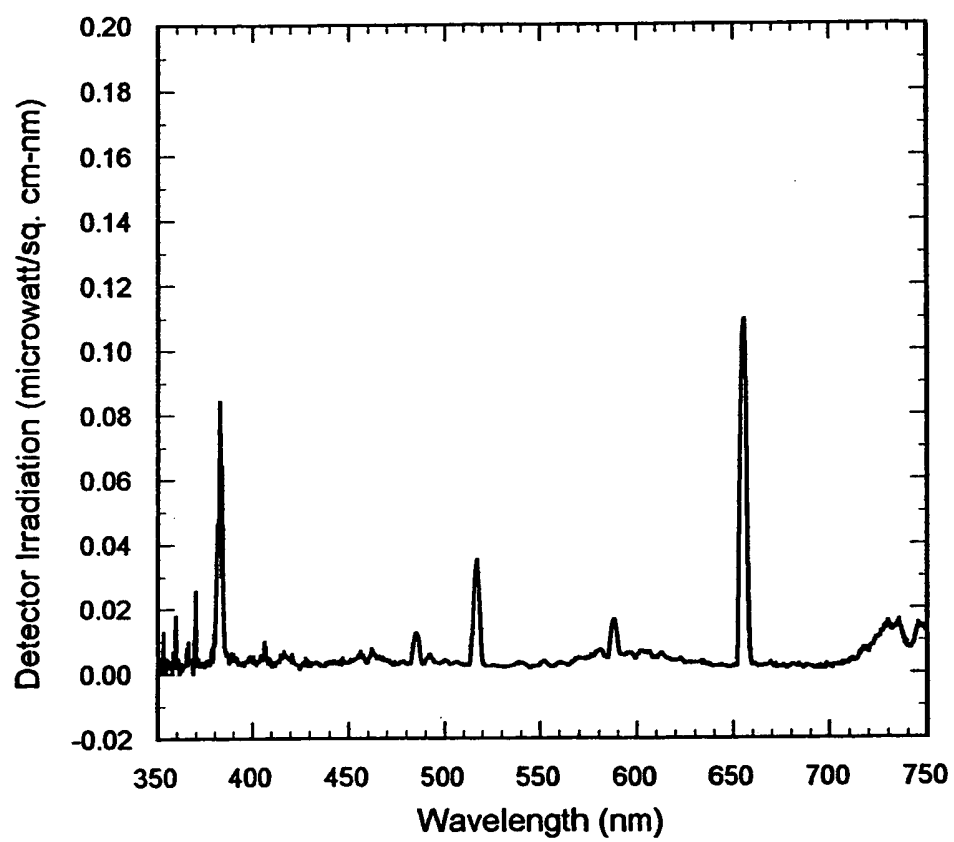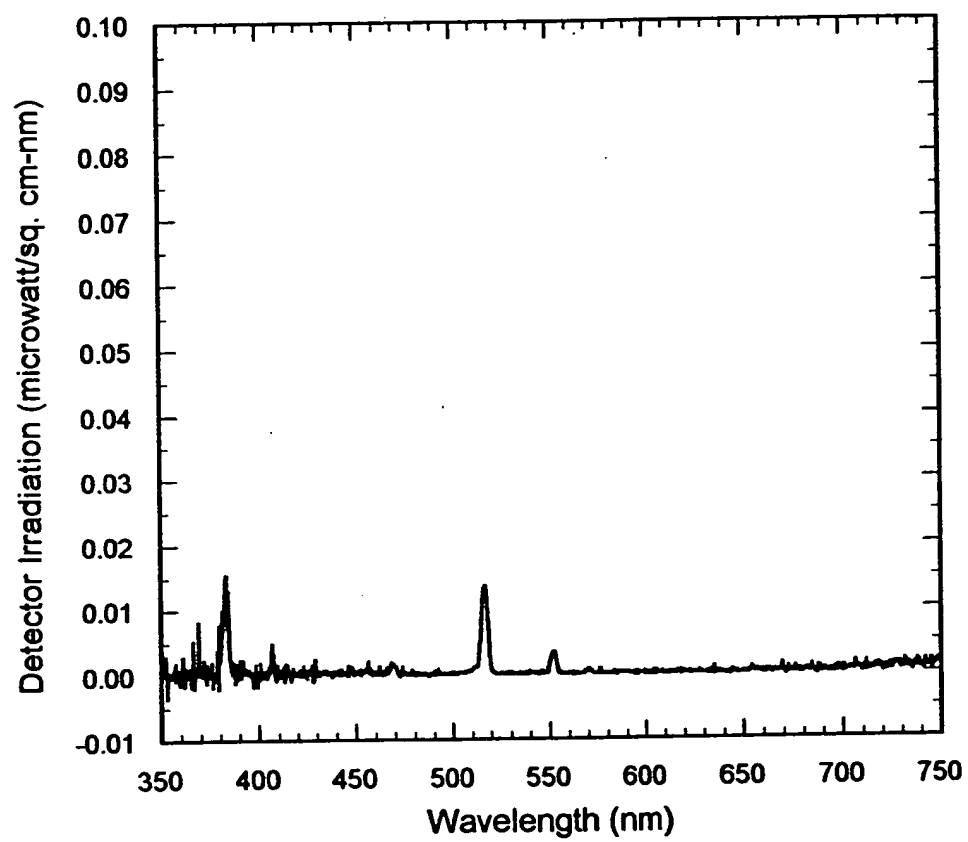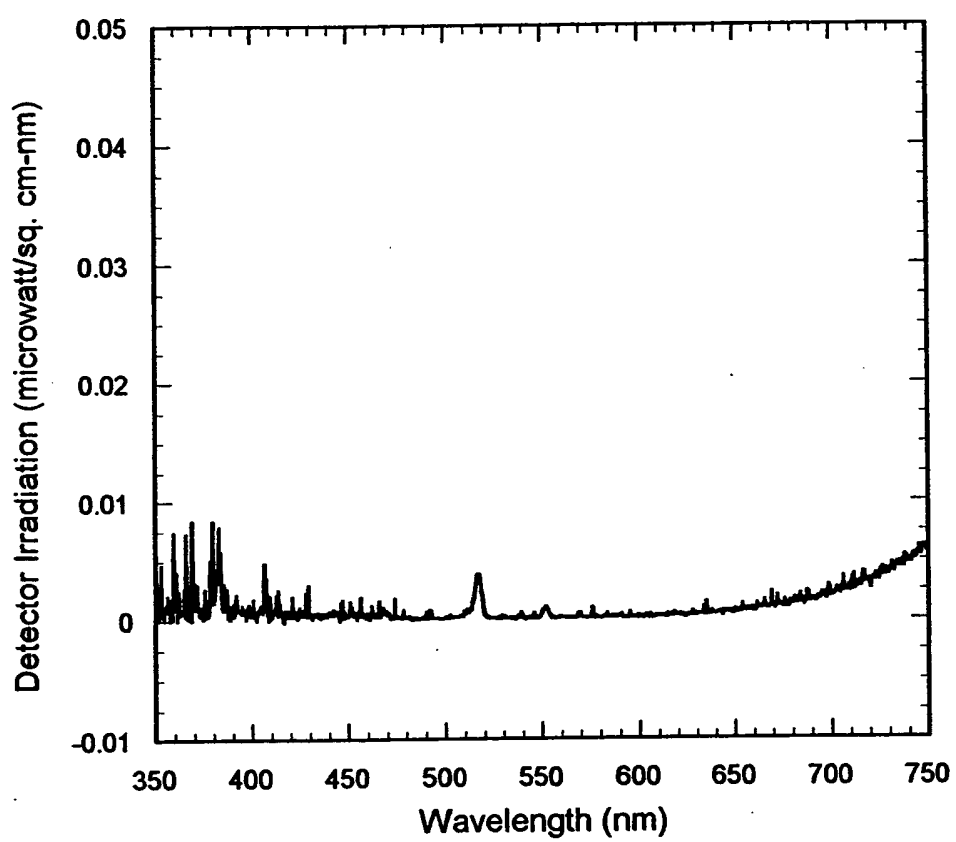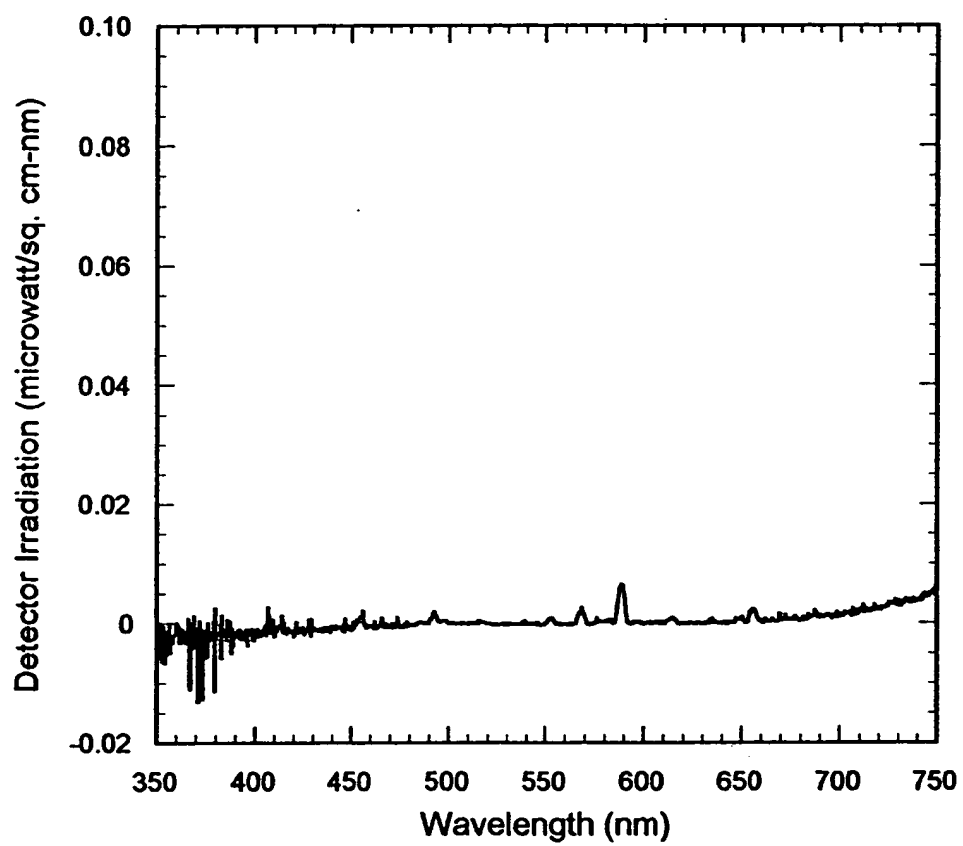
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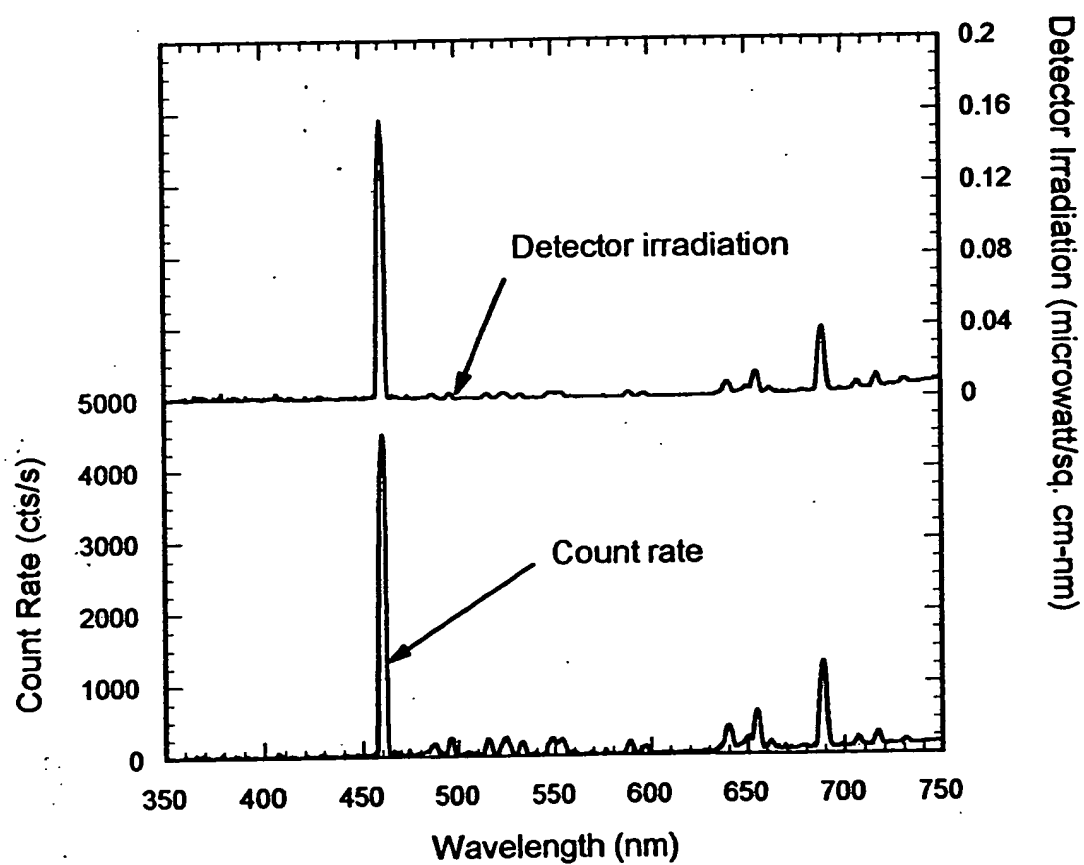
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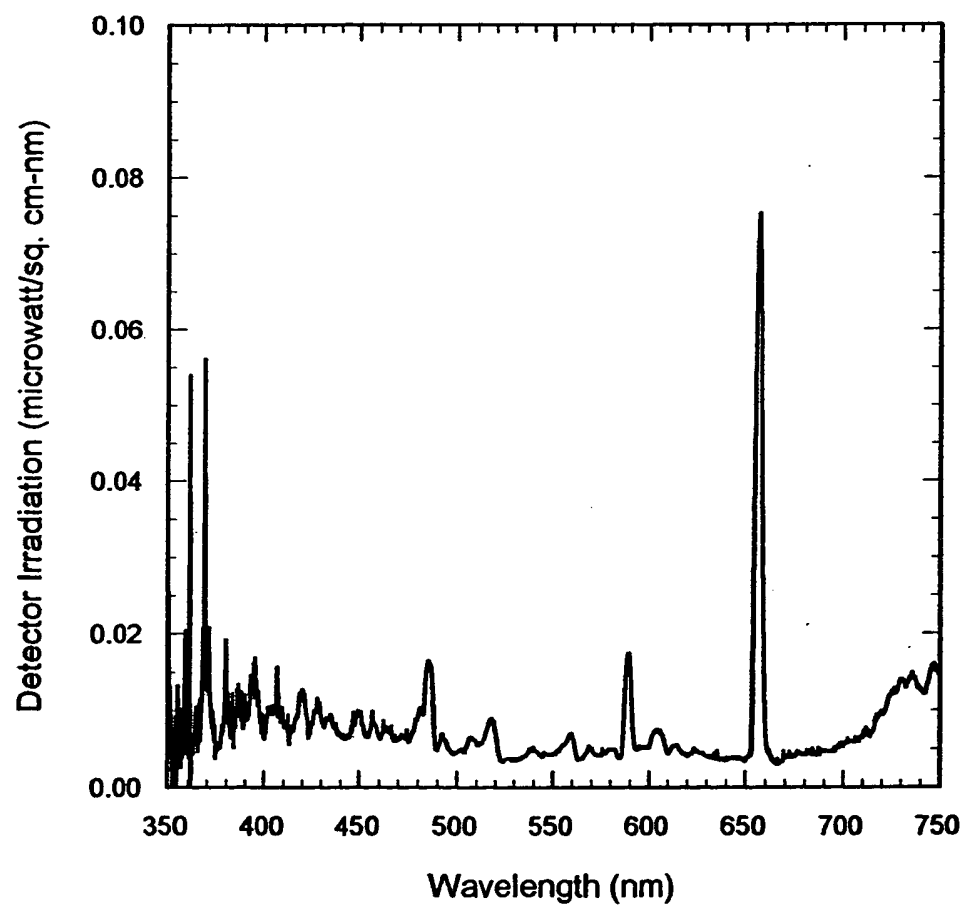
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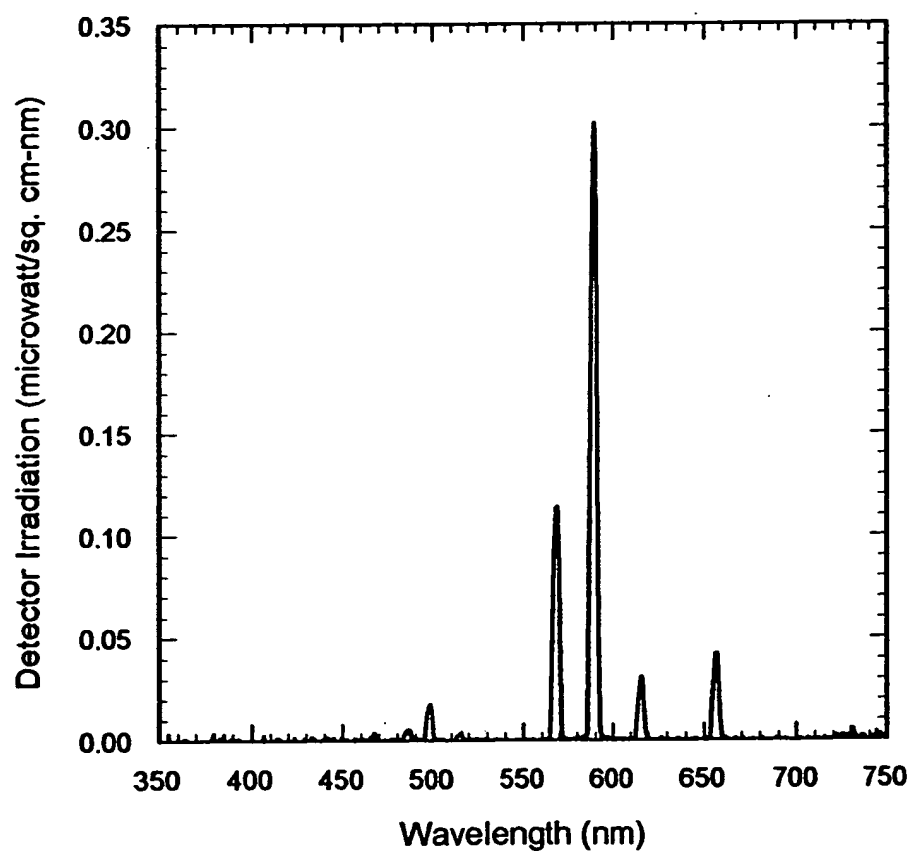
Figure 20

Figure 21

Figure 22

Figure 23

Figure 24

Figure 25

Figure 26

Figure 27

Figure 28